# Microwave Properties of High-$T_c$ Oxide Superconductors

G. Müller

*Fachbereich Physik der Bergischen Universität - Gesamthochschule Wuppertal*
*D-5600 Wuppertal 1, West Germany*

## INTRODUCTION

First results on the microwave properties of high-$T_c$ oxide superconductors have been reported two years ago at the last workshop on rf superconductivity by several groups [1-5]. In the meantime, improved samples of these promising materials have been investigated more systematically at various laboratories. In general, there are two different kinds of microwave measurements on high-$T_c$ superconductors. While the first ones using ESR apparatuses are focusing on their magnetic behaviour by means of uncalibrated microwave absorption studies, the second ones using specially designed set-ups are interested in absolute values of the surface impedance. Despite of the interesting information from the absorption studies about processes in granular superconductors at high magnetic field levels [6], in this article only the latter type of measurements are reviewed in order to judge about the applicability of high-$T_c$ oxide superconductors in microwave and electronic devices.

In the first chapter, some background information about the structural and transport properties of the oxide superconductors is summarized. Emphasis will be given to their highly anisotropic and granular features, which are most important for the observed microwave properties. Since misalignment of grains and phase impurities especially at grain boundaries limit the rf performance to a large extent, improved fabrication techniques for the synthesis of polycrystalline bulk ceramic and textured thick films as well as for singlecrystalline thin films will be shortly discussed in the next chapter. After a comparative description of various microwave measurement techniques developed for the investigation of samples of different shape and size, a survey of available microwave data for the oxide superconductors is given. In the main chapter, the most interesting results for the surface resistance as a function of temperature, frequency and rf magnetic field and for the temperature dependence of the penetration depth mainly of $YBa_2Cu_3O_7$ are presented and discussed. This will finally lead to conclusions about the present state of the art and future prospects for microwave and electronic applications of the high-$T_c$ oxide superconductors.

## STRUCTURAL AND TRANSPORT PROPERTIES

Since the discovery of oxide superconductors with a transition temperature $T_c$ above 30 K in 1986 [7], various oxide superconductors with perovskite ( $CaTiO_3$ )-like structure have been found until now [8]. The highest $T_c$ values occur for copper containing oxides, which can be devided into structural classes with equivalent combinations of elements. In Tab. 1, the most famous representants of six structural classes are listed together with their $T_c$ and with the range of $T_c$ values resulting for other members of the same class. These classes are usually called the doped 2:1:4 [7] and the strongly oxygen-deficient 1:2:3 [9, 10], 1:2:4 [11, 12], 2:2:0:1 [13], 2:2:1:2 [14, 15] and 2:2:2:3 [16] structures. Recently, a modified doped 2:1:4 structure with dominant electron instead of hole conduction have been found [17]. The main difference between all of these structures consists in the number n of four-fold planar-coordinated $CuO_2$ layers per unit cell, in which the normal and superconducting currents are believed to predominate and which seems to be roughly correlated with $T_c$. In Fig. 1, the crystal structure of $YBa_2Cu_3O_7$ having two of these layers is shown for example.

Tab. 1: Structural classes of copper-oxide superconductors with n $CuO_2$ layers.

| Most Famous Representant | $T_c$ | Range | n |
|---|---|---|---|
| $La_{1.85}Ba_{0.15}CuO_4$ | 35 K | 10–40 K | 1 |
| $YBa_2Cu_3O_7$ | 93 K | 60–96 K | 2 |
| $YBa_2Cu_4O_8$ | 81 K | 50–85 K | 2 |
| $Bi_2Sr_2CuO_6$ | 20 K | 10–90 K | 1 |
| $Bi_2Sr_2CaCu_2O_8$ | 83 K | < 110 K | 2 |
| $Tl_2Ba_2Ca_2Cu_3O_{10}$ | 122 K | < 125 K | 3 |

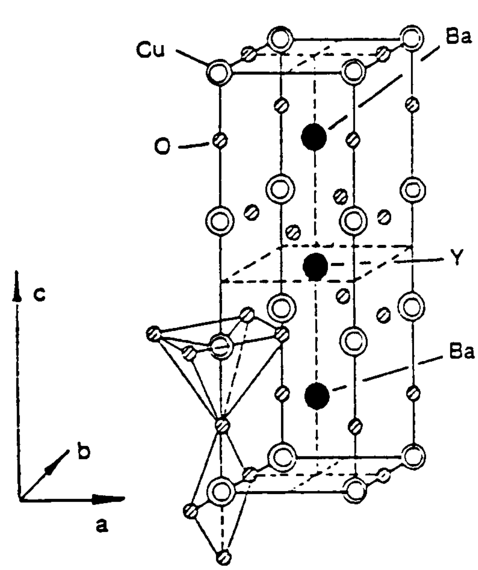



Fig. 1:
Crystal structure of $YBa_2Cu_3O_7$. The dimensions of the orthorhombic unit-cell are about a = 0.382 nm, b = 0.389 nm and c = 1.167 nm [10, 18].

The thermodynamic phase diagrams of these at least quaternary compounds and their subsystems are only partially known yet, but phase purity seems to be achievable only for some oxides of the 1:2:3 and of the 2:2:1:2 structure. Any remaining impurity phase like CuO, $BaCuO_2$ or $Y_2BaCuO_5$, however, will contribute to residual microwave losses. Moreover, the $T_c$ of the oxide superconductors depends sensitively on the oxygen content [19] and on metallic impurities [8]. Contaminations and atomic disorder at the surface should also enhance the surface resistance. Because of all these reasons, it is not surprising that sharp rf transitions have been obtained so far only for $YBa_2Cu_3O_7$ as we will see in the main chapter.

Beside these speculative disadvantages, there are two common features of all known oxide superconductors from which severe drawbacks concerning their applicability result. First of all, the layered structure causes strongly anisotropic transport properties of single crystals in the normal conducting [20] as well as in the superconducting state [21]. Secondly, the higher the $T_c$ the larger the energy gap $\Delta_0$ and the shorter the coherence length $\xi_0 = \hbar v_F / \pi \Delta_0$ ( $v_F$ = Fermi velocity ) of a superconductor is expected. Current estimates of $\xi_0$ for $YBa_2Cu_3O_7$ from dc magnetization measurements give anisotropic values of about 1-2 nm in the ab-plane but only 0.2-0.4 nm in the c-direction [22]. Since the latter value is of similar magnitude as the distance between the Cu-O layers, granular properties have to be considered for polycrystalline oxide superconductors [23]. Both features reduce the achievable critical current densities of $YBa_2Cu_3O_7$ wires [24] and exclude their application in magnets until now.

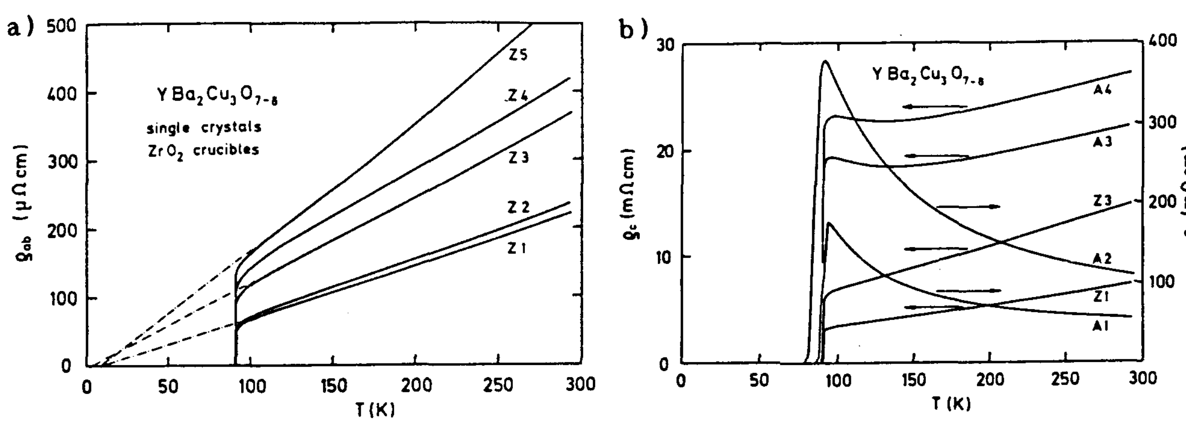



Fig. 2: Temperature dependence of the electrical resistivity of $YBa_2Cu_3O_7$ single crystals in the ab-plane ( a ) and in the c-direction ( b ) grown under different conditions in alumina ( A1 - A4 ) and zirconia ( Z1 - Z5 ) crucibles [25].

In Fig. 2 recent data on the anisotropic electrical resistivity $\rho(T)$ of $YBa_2Cu_3O_{7-\delta}$ are shown because of their importance for the surface impedance $Z_s$ in the normal state, which is given in the regime of the normal skin effect by [26]

$$Z_s = R_s + i\omega\mu_0\lambda = \rho/\delta\,(1+i) = \sqrt{\omega\mu_0\rho/2}\,(1+i) \qquad (1)$$

where $R_s$ is the surface resistance, $\omega = 2\pi f$ is the angular frequency and $\lambda = \delta/2$ is the field penetration depth which is correlated to the skin depth $\delta$. Obviously, both $\rho_{ab}$ and $\rho_c$ depend strongly on the quality of the sample. For the metal-like $\rho_{ab} \sim T$, the observed fluctuations are proposed to be caused by different carrier concentrations due to oxygen disorder in the ab-planes, while especially substitutional Al impurities seem to produce a semiconductor-like $\rho_c \sim 1/T$ [25]. Nevertheless, even for the best samples there remains a $\rho_{ab} \approx 60\,\mu\Omega$cm at 100 K, which is very high compared to that of copper, and an intrinsic anisotropy of $\rho_c/\rho_{ab}$ of about 60. According to (1), at 10 GHz and 100 K values for $R_s$ between 0.15 $\Omega$ and 1.2 $\Omega$ and for $\delta$ between 3.9 $\mu$m and 30 $\mu$m result depending on the orientation of the surface currents, respectively. Therefore, rather thick layers are needed to measure $Z_s$ of the oxide superconductors above $T_c$ correctly.

Another important parameter for microwave applications at high field levels, which should be also anisotropic, is the thermal conductivity. However, until now only mean values for polycrystalline bulk samples of $YBa_2Cu_3O_{7-\delta}$ have been measured as shown in Fig. 3. The peak at 60 K indicates dominant phonon heat transport at all temperatures below $T_c$ [27]. Moreover, porosity can reduce the whole $\lambda(T)$ curve by about one order of magnitude [27, 28]. Comparing the absolute values with those of Nb and $Nb_3Sn$ and remembering local thermal instabilities in such cavities with low thermal conductivity walls [29-31], thin films of some $\mu$m in thickness on substrates with high thermal conductivity will be required for the thermal stabilization of indirectly cooled oxide superconductors at high rf magnetic fields.

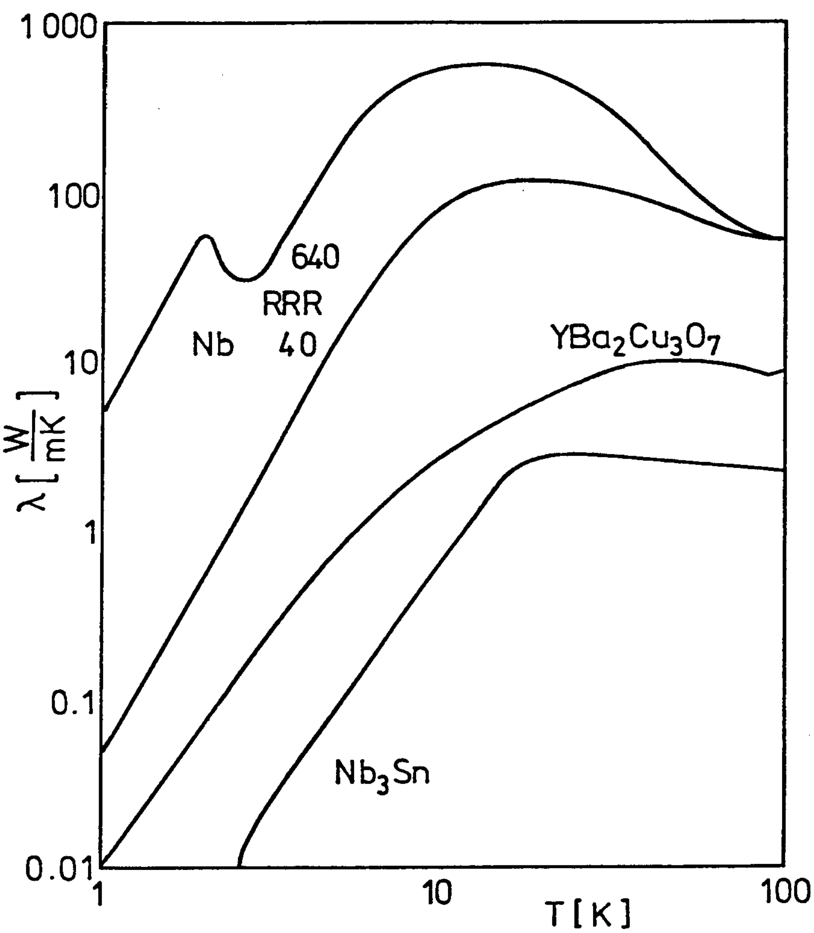



Fig. 3: Temperature dependence of the thermal conductivity $\lambda(T)$ of polycrystalline $YBa_2Cu_3O_{7-\delta}$ [27] in comparison with that of Nb [30, 32] and $Nb_3Sn$ [33].

Tab. 2: Comparison of material parameters of superconductors which are important for microwave applications.

| Material | $T_c$ [K] | $\Delta_0/kT_c$ | $2\Delta_0/h$ [THz] | $\xi_0(0)$ [nm] | $\lambda(0)$ [nm] | $\mu_0 H_c(0)$ [T] | $\mu_0 H_{sh}(0)$ [T] |
|---|---|---|---|---|---|---|---|
| Pb | 7.2 | 2.17 | 0.65 | 83 | 48 | 0.08 | 0.12 |
| Nb | 9.2 | 1.97 | 0.75 | 39 | 40 | 0.20 | 0.21 |
| $Nb_3Sn$ | 18 | 2.20 | 1.65 | 5.7 | 110 | 0.535 | 0.40 |
| $YBa_2Cu_3O_7$ (preliminary) | 93 | 1.5–4 (anisotropic?) | 6–15 | ab: < 2<br>c: < 0.4 | $l_{ab}$: 140<br>$l_c$: 770 | 1.0–1.4 | 0.75–1.05 |

In Tab. 2, preliminary values for some basic material parameters of superconducting $YBa_2Cu_3O_7$ are compared to the much better known ones of Pb, Nb and $Nb_3Sn$. The quoted range of values for the reduced energy gap $\Delta_0/kT_c$ and the gap frequency $2\Delta_0/h$ corresponds to actual reviews about tunneling [34], infrared [35] and photoemission [36] measurements, which are still controversial concerning a possible anisotropy of $\Delta_0$. The magnetic field penetration depth $\lambda(0)$ has been determined on single crystals from muon-spin-rotation [37] and dc magnetization [38] measurements consistently to be about 140 nm for currents in the ab-plane, while the best evidence for the anisotropy of $\lambda_a : \lambda_b : \lambda_c = 1.2 : 1 : 5.5$ result from vortex-lattice decoration experiments [39]. The thermodynamic critical field $H_c(0)$ has been estimated from a review of specific heat measurements [40]. Since the oxide superconductors are in the extreme type II regime, the resulting intrinsic critical rf magnetic field will be given by the superheating field $H_{sh} = 0.75\, H_c$ [41]. In summary, the high-$T_c$ oxide superconductors are promising for electronic and microwave devices not only because of practical cryogenic reasons but also because of their potential extension to higher frequencies and field levels of operation. As an example, accelerator cavities with a maximum accelerating field of 250 MV/m, which corresponds typically to a peak magnetic surface field of 1 T, can be envisaged.

Such a proposal would be unrealistic, however, as long as the granular properties of the oxide superconductors cannot be eliminated. Recently, a phenomenological theory for anisotropic granular superconductors has been developed [42]. Based on a network of well-oriented Josephson-coupled grains ( see Fig. 4 ), simple formulas for the effective resistivity and penetration depth of the oxide superconductors have been derived. The main ingredients of this theory are a straightforward London and Ginzburg-Landau theory with an anisotropic effective mass tensor of the charge carriers and anisotropic intergranular Josephson currents $I_i = I_{i0} \sin\Delta\gamma_i$, which oscillate in the i-direction with the phase difference $\Delta\gamma_i$ of the wave functions across the junction. The maximum Josephson current $I_{i0}$ is much smaller than the intragrain current limit and is given by [43]

$$I_{i0}(T) = \frac{\pi\,\Delta(T)}{2\,e\,R_{in}} \tanh\frac{\Delta(T)}{2\,k\,T} \qquad (2)$$

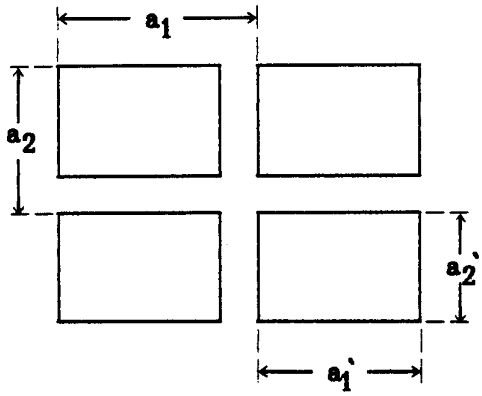



Fig. 4:
Model of Josephson-coupled blocks of size $a_i'$ in a periodic array with lattice parameters $a_i$ [42]. In the three dimensional model, the area of the junction in i-direction is $A_i' = a_j' a_k'$ and of the unit cell is $A_i = a_j a_k$.

where $R_{in}$ is the normal-state tunneling resistance across the junction. As the result of this theory, the intrinsic intragranular values of the normal-state resistivity $\rho_{i0}$ and of the penetration depth $\lambda_{i0}$ become somewhat enlarged to the following effective values:

$$\rho_i = \rho_{i0} \frac{a_i' A_i}{a_i A_i'} + R_{in} \frac{A_i}{a_i} \qquad (3)$$

$$\lambda_i^2 = \lambda_{i0}^2 \frac{a_i' A_i}{a_i A_i'} + \frac{\hbar}{2 e \mu_0 I_{i0}} \frac{A_i}{a_i} \qquad (4)$$

In the limiting case of strong Josephson-coupling between the grains, $\rho_i$ and $\lambda_i$ are dominated of course by the intrinsic values and remain independent quantities. In the weak Josephson-coupling limit, however, the terms with the junction parameters $R_{in}$ and $I_{i0}$ become dominant for $\rho_i$ and $\lambda_i$ leading with (2) to the correlation:

$$\lambda_i^2 = \rho_i \frac{\hbar}{\pi \mu_0 \Delta(T)} \coth\frac{\Delta(T)}{2 k T} \qquad (5)$$

If there is an intrinsic granularity of the high-$T_c$ supercondcutors due to weak coupling in the c-direction, a corresponding lower value of an anisotropic energy gap $\Delta(0)$ of $YBa_2Cu_3O_7$ can be estimated from (5) with measured $\lambda_c(0)$ (Tab. 2) and extrapolated $\rho_c(0)$ (Fig. 2) values to be in the order of 7 meV ($\cong 0.9\,kT_c$). In any case, inhomogeneities of real materials will cause currents to flow perpendicular to the surface, thereby increasing $\lambda$ [44] and possibly introducing granular behaviour even for highly c-axis oriented thin films.

Despite of the unsolved questions about the coupling mechanism and the calculation of the intrinsic surface impedance of the oxide superconductors, the additional term for $R_s$ resulting for granular superconductors can be derived as well as for $\lambda$ [45]

$$R_s = \omega^2 \frac{\mu_0 \hbar}{4 e I_{i0} R_{in}} \frac{\lambda_j^2}{\lambda} \qquad (6)$$

where $\lambda_j^2$ is the second term in (4). In the weak-coupling limit $\lambda$ approaches $\lambda_j$, and replacing $I_{i0} R_{in}$ by $\Delta$ according to (2) leads to a simple correlation between the residual surface resistance $R_s(0)$ and $\lambda(0)$:

$$R_s(0) = \omega^2 \frac{\mu_0 \hbar}{2 \pi \Delta(0)} \lambda(0) \qquad (7)$$

Therefore, assuming for $YBa_2Cu_3O_7$ the same parameters as above, an intrinsic strong granularity due to weak-coupling in the c-direction should limit the residual surface resistance to values in the order of

$$R_s(0) = 0.57\,\mu\Omega \cdot f\,[\mathrm{GHz}]^2 \qquad (8)$$

These considerations are restricted, however, to low magnetic field levels because of their strong influence on the weak links. Above a threshold surface field $H_{ij}$ which corresponds to the critical Josephson current $I_{i0}$ across the junctions, decoupling of the grains leads to additional microwave losses. Similar to the ac losses in the critical flux state, these losses should increase linearly both with f and with the magnetic field $H_s$ applied to the surface [6, 46]:

$$\frac{R_s}{H_s} = \frac{4\,\mu_0 f}{3\,J_c} = \frac{0.17\ f\,[\mathrm{GHz}]}{J_c\,[\mathrm{A/cm2}]}\ \frac{\Omega}{\mathrm{A/m}} \qquad (9)$$

For typical values of the critical current density $J_c$ of polycrystalline $YBa_2Cu_3O_{7-\delta}$ in the order of $10^3$ $A/cm^2$ [24] and for 1 GHz, the resulting $R_s$ exceeds 1 m$\Omega$ at a $H_s$ of about 6 A/m ($\hat{=}$ 7.54 $\mu$T). Therefore, oxide superconductors with low $J_c$ limited by weak links seem to be suited neither for magnet nor for accelerator applications.

## SYNTHESIS OF BULK, THICK AND THIN FILMS

From the previous chapter it has become evident that a careful synthesis of the oxide superconductors is required for good microwave properties. Therefore, well approved fabrication techniques for polycrystalline bulk ceramic and thick layers as well as for singlecrystalline thin films shall be mentioned here. The details of these descriptions are restricted exemplarily to $YBa_2Cu_3O_{7-\delta}$ , which has been investigated most, but the basic considerations can be applied to all oxide superconductors. The quality of the resultant samples will be compared by standard characterization techniques like scanning electron microscopy, X-ray diffractometry and resistively as well as inductively measured transition curves and critical current densities.

Polycrystalline high-$T_c$ superconductors are usually synthesized from metal oxides and carbonates by solid-state reaction at high temperatures. In case of $YBa_2Cu_3O_{7-\delta}$, at first a stoichiometric mixture of dried $Y_2O_3$, $BaCO_3$ and CuO of high purity ($\geq$ 99.99 %) and small grain size ($\leq$ 10 $\mu$m) is ball milled for about one hour. Then the calcination is performed in air or oxygen at temperatures between 910$^o$C and 930$^o$C for about 100 h. Repeated interruption of the calcination for additional ball milling of the agglomerate improves the homogeneity of the superconducting powder significantly [47]. After the final ball milling, an average particle size of typically 5 $\mu$m results, which can be further reduced with sieves or by sedimentation in organic liquids to less than 1 $\mu$m. Care must be taken of contaminations from mechanical abrasion or chemical reaction, respectively. Bulk ceramic pellets or rings are pressed directly from this powder, while for wire or thick layer fabrication organic additives are necessary. Finally, these samples are sintered at 900$^o$C in air or at 920$^o$C in pure oxygen for extended time periods up to 2 weeks. For these sintering conditions, a sufficient grain growth with a minimum of impurity phases like CuO, $BaCuO_2$ and $Y_2BaCuO_5$ is achieved [48]. The phase transition from the insulating tetragonal ( $\delta = 1$ ) to the superconducting orthorhombic ( $\delta = 0$ ) structure at temperatures near 700°C [49] can be completed in an $O_2$ atmosphere during the naturally slow ( ≥ 4 h ) cool-down of the furnace. After the shrinkage during the sintering, more than 90 % of the theoretical density ( 6.3 g/cm$^3$ ) is achievable for long-annealed bulk ceramic $YBa_2Cu_3O_{7-\delta}$ samples.

For large microwave devices of complex shape like cavities, the coating of mechnically rigid substrates with thick polycrystalline films of oxide superconductors is desirable. Among the various coating techniques developed for this purpose, the electrophoretic deposition of thick $YBa_2Cu_3O_{7-\delta}$ films onto silver substrates [50] has given the best results. Electrophoresis describes the migration of charged microscopic particles through a stationary liquid under the influence of an electrostatic field. In an organic suspension from calcined $YBa_2Cu_3O_{7-\delta}$ powder and n-butanol, for example, a net positive charging of the colloids leads to a cataphoresis as shown in Fig. 5. The quality of the resultant film can be improved by restriction on the smallest ( < 2 μm ) particles due to sedimentation for about 10 h. Such a suspension with remaining 10 g/l leads at 45°C and 180 V/m typically to a current density of 30 μA/cm$^2$ and to a 5 μm thick layer within two minutes. Moreover, a high degree of c-axis texturing perpendicular to the film surface can be achieved if the deposition is performed in a strong magnetostatic field of 8 T [51, 52]. Because of the shrinkage during sintering, stepwise deposition followed by short drying and sintering for half an hour at 920°C is needed to obtain homogenous films of about 20 μm in thickness, which are finally sintered under the same conditions as bulk samples.

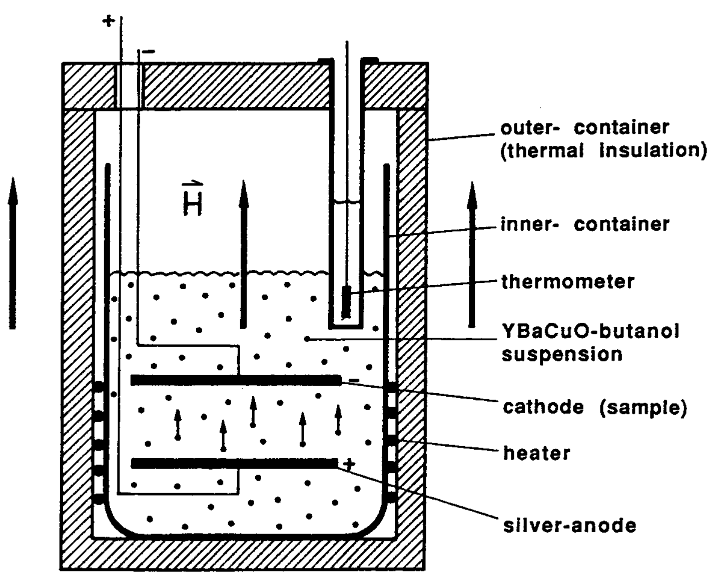



Fig. 5:
Schematic setup for the electrophoretic deposition of textured layers of oxide superconductors onto metallic substrates [51].

For planar microwave and electronic devices, epitaxially grown thin films on single-crystalline dielectric substrates are most promising. The choice of suitable substrates underlies three main requirements. First, their crystal structure has to match closely that of the oxide superconductors to support epitaxial growth with c-axis perpendicular to the surface. Secondly, their dielectric loss tangent has to be as small as possible to avoid additional microwave losses which limit the performance of superconducting devices. Last but not least, the substrates have to withstand the elevated temperatures for the formation of oxide superconductors without too much interdiffusion, which can lead to reduced film quality and enhanced dielectric losses. While the substrates listed in Tab. 3 are more or less all approved concerning the first and third issue, the second

| | MgO | $ZrO_2$ | $SrTiO_3$ | $LaGaO_3$ | $LaAlO_3$ |
|---|---|---|---|---|---|
| $\varepsilon_r(T)$ | 10 | 25 | $10^2$-$10^4$ | 25 | 16 |
| $\tan\delta(T)$ | $\leq 10^{-3}$ | $\leq 10^{-2}$ | $10^{-2}$ | $10^{-3}$-$10^{-6}$ | $10^{-3}$-$10^{-6}$ |

Tab. 3:
Permittivity $\varepsilon_r$ and typical $\tan\delta$ of dielectric substrates approved for $YBa_2Cu_3O_7$ thin films at $T \leq 300$ K and microwave frequencies [53].

requirement has enforced the search for alternative materials with much improved microwave properties like $LaGaO_3$ [54] and $LaAlO_3$ [55] or for buffer layers on $Al_2O_3$ [53], which provides very low $\tan\delta$ values but poisons $YBa_2Cu_3O_7$. Moreover, the quality of the thin films depends mainly on the in situ crystalline growth conditions, i.e. on the chosen combination of substrate temperature and oxygen pressure [56], but less on the specially used physical or chemical mass transport process as long as the correct metallic stoichiometry is supplied. Deposition techniques which cannot be performed under sufficient high oxygen pressure, however, require a post annealing of the film in pure oxygen at temperatures around 900°C resulting in reduced film quality [57]. The best results have been achieved so far with the so-called laser ablation technique [58, 59]. In this easily reproducible and fast process, stoichiometric superconductor targets are ablated by an UV excimer laser of short pulse length ( $\leq$ 60 ns ) but high pulse energy ( $\leq$ 2 J ) and repetition rate ( $\leq$ 100 Hz ) as shown in Fig. 6. At typical operating parameters of 2 J/cm$^2$ energy density, 0.2 mbar $O_2$ pressure and 750°C substrate temperature, film growth rates up to 0.1 μm per minute are obtained on 10x10 mm$^2$ substrates. Scanning systems will be necessary for larger samples. Alternatively, the much slower sputtering techniques can be also applied for large scale in situ film growth [57].

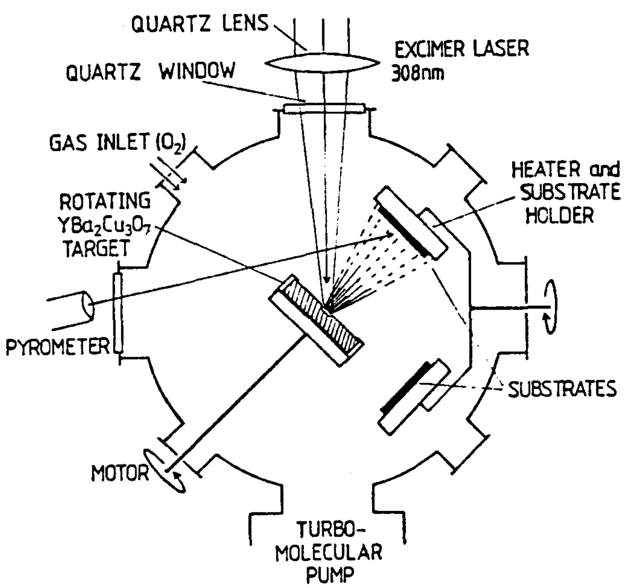
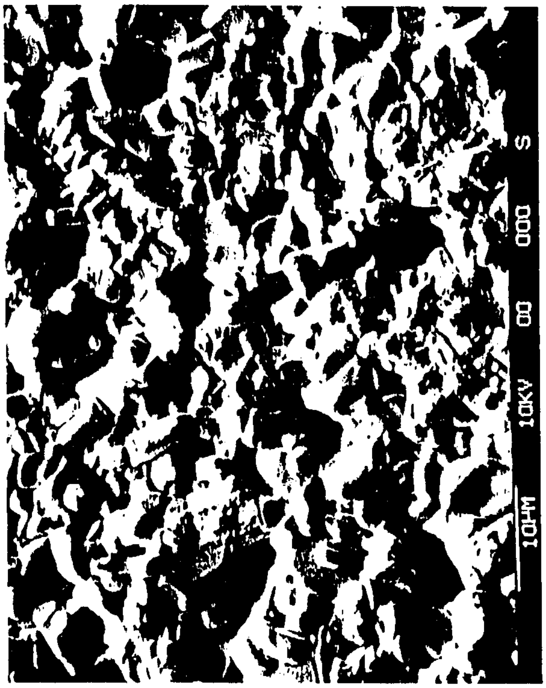



Fig. 6:
Schematic setup for the deposition of singlecrystalline oxide superconductors by laser ablation from stoichiometric targets [59].

The surface quality of $YBa_2Cu_3O_{7-\delta}$ samples resulting for these different techniques is compared in Fig. 7. While polycrystalline bulk ceramic and untextured thick films provide a porous surface with well separated grains ( Fig. 7a ), c-axis texturing leads to a platelet structure due to the merging of grains within the ab-plane ( Fig. 7b ). In contrast, singlecrystalline films exhibit a rather smooth surface often with particulate inhomogeneities of μm size ( Fig. 7c ). Nevertheless, microprobe analysis yields within ± 1 % the correct stoichiometry for at least 95 % of the surface of all these samples.

a)



b)

c)

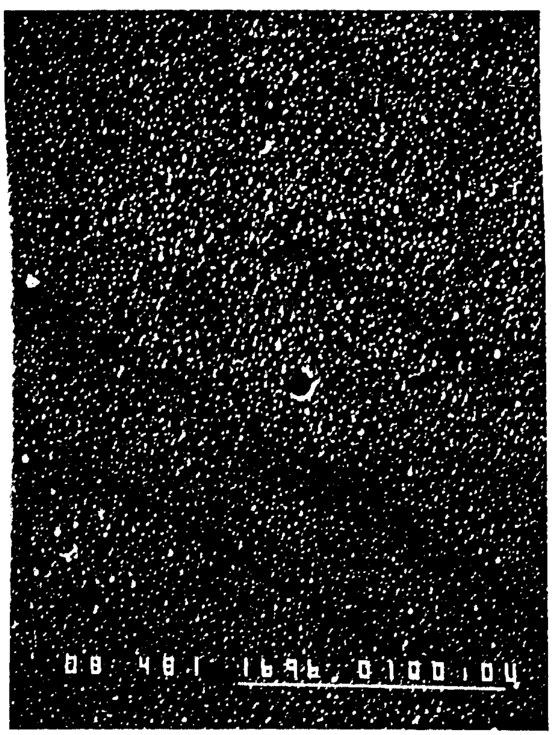
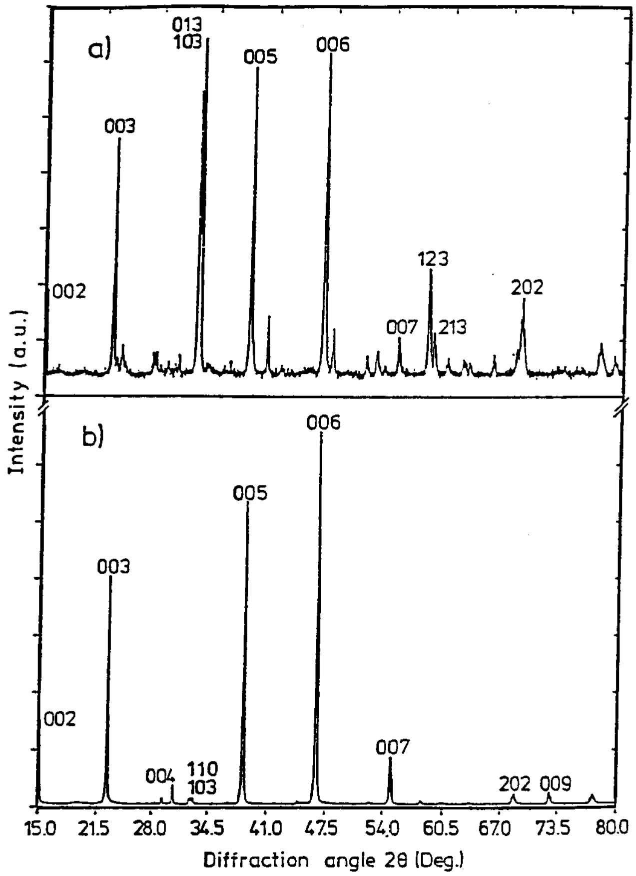


Fig. 7:
Scanning electron micrographs of the surface of an untextured (a) and a c-axis textured electrophoretically deposited thick film (b) [51] and a laser ablazed thin film (c) [60] of $YBa_2Cu_3O_{7-\delta}$.

The X-ray diffraction patterns of the same $YBa_2Cu_3O_{7-\delta}$ samples are displayed in Fig. 8. In comparison, bulk ceramic and untextured thick films show much more peaks and larger background intensity than c-axis textured thick and thin films. It is remarkable that the dominant 013/103 peak typical for polycrystalline $YBa_2Cu_3O_7$ can be suppressed by about a factor of 50 for the thick film just by the alignment of the particles during the electrophoresis in a strong magnetic field. However, only the singlecrystalline thin film exhibits a clear pattern of sharp (00n) lines typical for complete c-axis orientation. Moreover, all three patterns confirm an extensive phase purity of the samples.



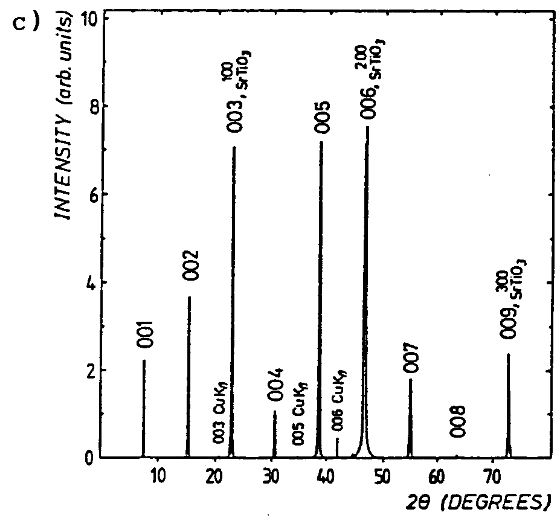




Fig. 8:
X-ray diffraction patterns for the same $YBa_2Cu_3O_{7-\delta}$ samples (a - c) as in Fig. 7.

It is well known that inductively measured transition curves provide a much deeper insight into the quality of samples than resistively measured ones. In inhomogenous or granular material, a percolative superconducting path causes zero resistance already, while impurities and weak links lead to an incomplete Meißner effect and to a broadened inductive transition with reduced $T_c$ especially in high magnetic fields [24]. Accordingly,

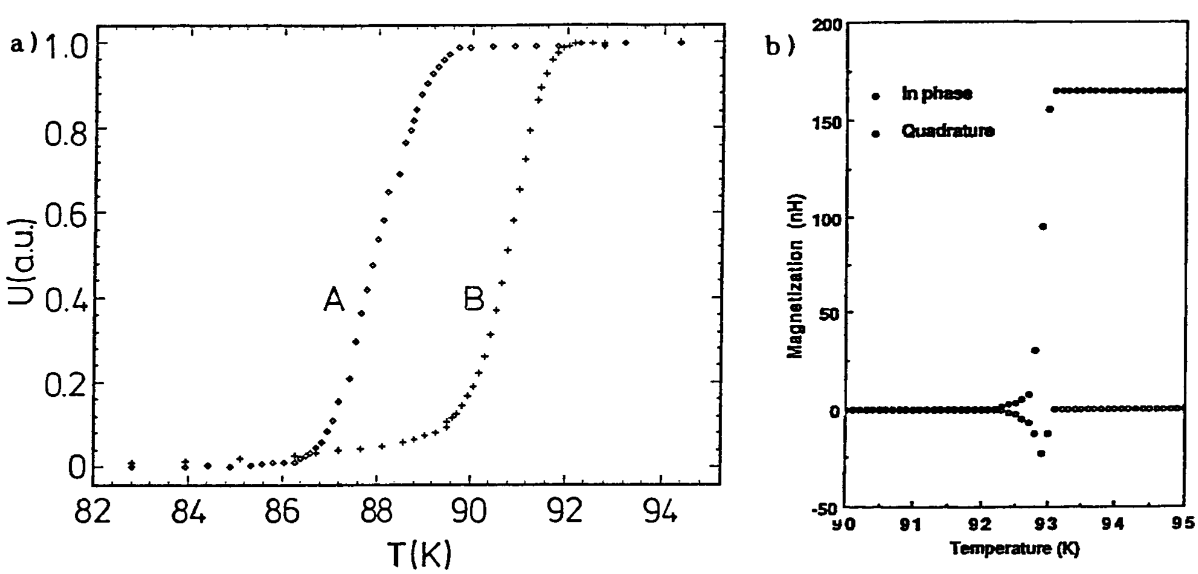



Fig. 9: Comparison of inductively measured transition curves of a) electrophoretically deposited polycrystalline thick films without (A) and with (B) c-axis texturing [51] and b) laser ablazed singlecrystalline thin films [61] of $YBa_2Cu_3O_{7-\delta}$.

there is a significant difference in the magnetic shielding behaviour of bulk ceramic or untextured thick films and c-axis textured ones as shown in Fig. 9a. Despite of similar resistive $T_c$ values above 92 K, a significant inductive $T_c$ shift from 88 K for the untextured to 91 K for the textured thick film results due to the improved coupling between the grains in the ab-plane [51]. Still narrower inductive transitions close to the optimum $T_c$ of 93 K have been obtained for epitaxially grown $YBa_2Cu_3O_{7-\delta}$ thin films on $LaAlO_3$ (see Fig. 9b). These observations can be explained by the distinction between intergranular and intragrain currents [23] which are both obliged to contribute to macroscopic shielding currents. Direct measurements of the critical current density within single-crystalline thin films [58, 59] and across single grain boundaries [62] have given at 4.2 K more than $10^7$ A/cm$^2$ for intragrain currents in favourable directions but only less than $10^4$ A/cm2 for intergranular currents, both of which are further reduced by magnetic fields. The resultant $J_c$ of polycrystalline material stays at 77 K and low magnetic fields below $10^3$ A/cm$^2$.

## MICROWAVE MEASUREMENT TECHNIQUES

There are two important differences between dc or ac and microwave measurements. First, all microwave losses are produced only in a very thin surface layer in the order of magnitude of the magnetic field penetration depth $\lambda$ (see Tab. 2). For homogenous superconductors with a small coherence length, $\lambda$ is expected to be equal to the London penetration depth [63], while for granular superconductors the effective $\lambda$ (4) should be enlarged. Secondly, the shielding currents are forced to flow everywhere in this surface layer, i.e. any remaining unpaired conduction electron will convert microwave into thermal energy. Therefore, even homogenous superconductors provide losses due to thermally broken electron pairs, the density of which is correlated to the energy gap. In a simple two fluid model, scaling laws for the frequency and temperature dependence of the intrinsic penetration depth and surface resistance can be derived [64]

$$\lambda(T) = \lambda(0)/\sqrt{1 - t^4} \qquad (10)$$

$$R_s(\omega, T) \sim \omega^2 t^4 (1 - t^2)/(1 - t^4)^2 \qquad \text{with } t = T/T_c \qquad (11)$$

which have proven to be near $T_c$ good approximations for all classical superconductors. At temperatures below 0.5 $T_c$, the existence of an energy gap $\Delta_0$ leads to the modified temperature dependencies [50]

$$\lambda(T) - \lambda(0) \sim e^{-\Delta_0/kT} \qquad (12)$$

$$R_s(\omega, T) = A\,\omega^2/T\; e^{-\Delta_0/kT} \qquad (13)$$

Both formulas describe measured data of classical superconductors quite well as long as an additional temperature independent residual surface resistance $R_{res}$ is introduced, which accounts for surface impurities and other imperfections like frozen-in magnetic flux or grain boundaries as discussed before (7). For the absolute calculation of the surface impedance of weak-coupling superconductors in the whole temperature range, computer programs based on the BCS theory have been written [65, 66].

For the determination of the surface impedance of oxide superconductors, various measurement techniques are used which are optimized for different shapes and sizes of samples. The very first experiments with small ceramic pellets of $YBa_2Cu_3O_{7-\delta}$ were performed in a rather simple way by putting them into a niobium host cavity [67]. This method provides information about $R_S(T)$ and $\lambda(T)$ above and just below $T_C$, and also at temperatures below 4.2 K as long as the losses in the sample dominate the total losses. Additional irreproducible losses due to contact currents have been successfully avoided by an insulating $Nb_2O_5$ layer [2]. A much improved technique which allows accurate $Z_S$ measurements in the whole temperature range has been developed at Northeastern [1, 68] and Cornell [3, 69] Universities. The main idea is to mount the sample on an thermally insulated sapphire rod as shown in Fig. 10, so that its temperature can be varied while the host cavity remains superconducting at 4.2 K. Meanwhile, similar cavities are in operation also at other laboratories [70-72]. The useful frequency range for both types of host cavities is restricted to about 1 - 20 GHz because of sensitivity and size reasons, respectively. Such apparatuses are well suited for the investigation of single crystals of complicate shape and for small samples of singlecrystalline thin films on low-loss dielectric substrates, if they are calibrated properly. Moreover, they allow to study the dependence of $R_S$ on the rf magnetic field strength up to the limit set by the superconducting host cavity.

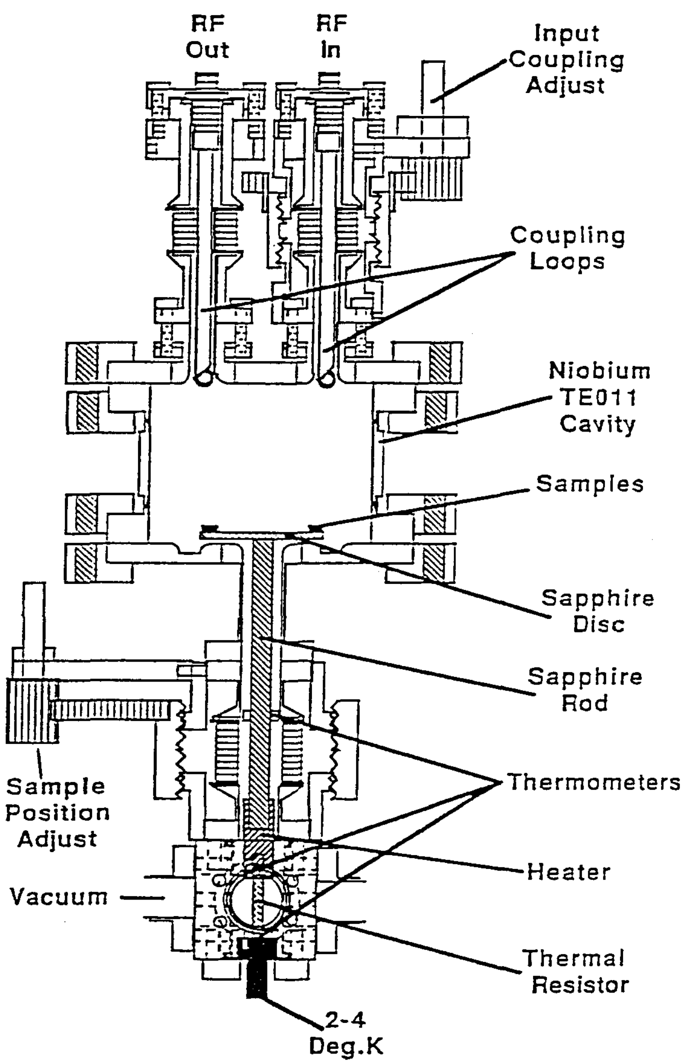



Fig. 10:
Cylindrical Nb cavity for the test of small samples on an independently heated sapphire support in the $TE_{011}$ mode at 6 GHz [69].

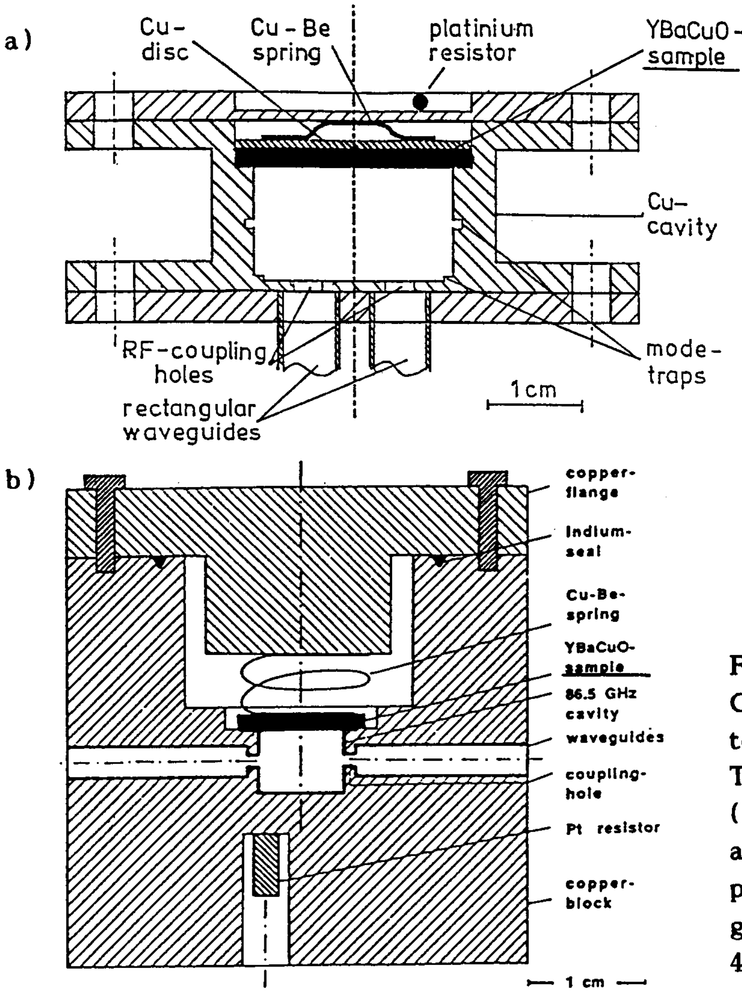



Fig. 11:
Cylindrical copper cavities for the test of plane samples (a) in the $TE_{011}$ mode at 21.4 GHz [47] and (b) in the $TE_{013}$ and $TE_{021}$ mode at about 87 GHz [60]. The corresponding ratio of total to partial geometric factor amounts 24 %, 40 % and 5 %, respectively.

A much better defined test geometry can be realized with plane samples of any size by replacing one endplate of copper or niobium cavities as shown in Fig. 11. By means of its partial geometric factor, which is calculable from the field configuration of the testing mode, $R_s(T)$ and $\lambda(T)$ result from two consecutive measurements of the quality factor and the resonant frequency shift with and without the sample [50]. Such pill-box cavities are usually designed for operation in the $TE_{0np}$ mode family to avoid currents across the contact surface. The degeneracy with the $TM_{1np}$ modes has to be splitted sufficiently by mode traps. The main disadvantage of this measurement technique consists in the common temperature of sample and cavity, which limits the sensitivity for the losses in the sample to about 10 % of the total losses. Therefore, OFHC copper cavities are preferred in the transition regime, while superconducting niobium cavities enable higher accuracy and field levels only at temperatures below 4.2 K. Nevertheless, similar cavities are used in most of the microwave laboratories. Two modifications have been pursued to extend the bandwidth of such measurements. One is a movable piston for a variable resonance frequency of the testing mode [73], and the other is a dielectric loading of the cavity with sapphire for lower operating frequency with small samples [74]. Moreover, for the investigation of two plane samples a post dielectric resonator without any side wall is sufficient [75]. For very thin films of oxide superconductors, however, transmission losses must be taken into account to extract the true $R_s$ and $\lambda$ values from the measured surface impedance [76]. Alternatively, the transmission loss of such films can be measured directly in a non-resonant way [77].

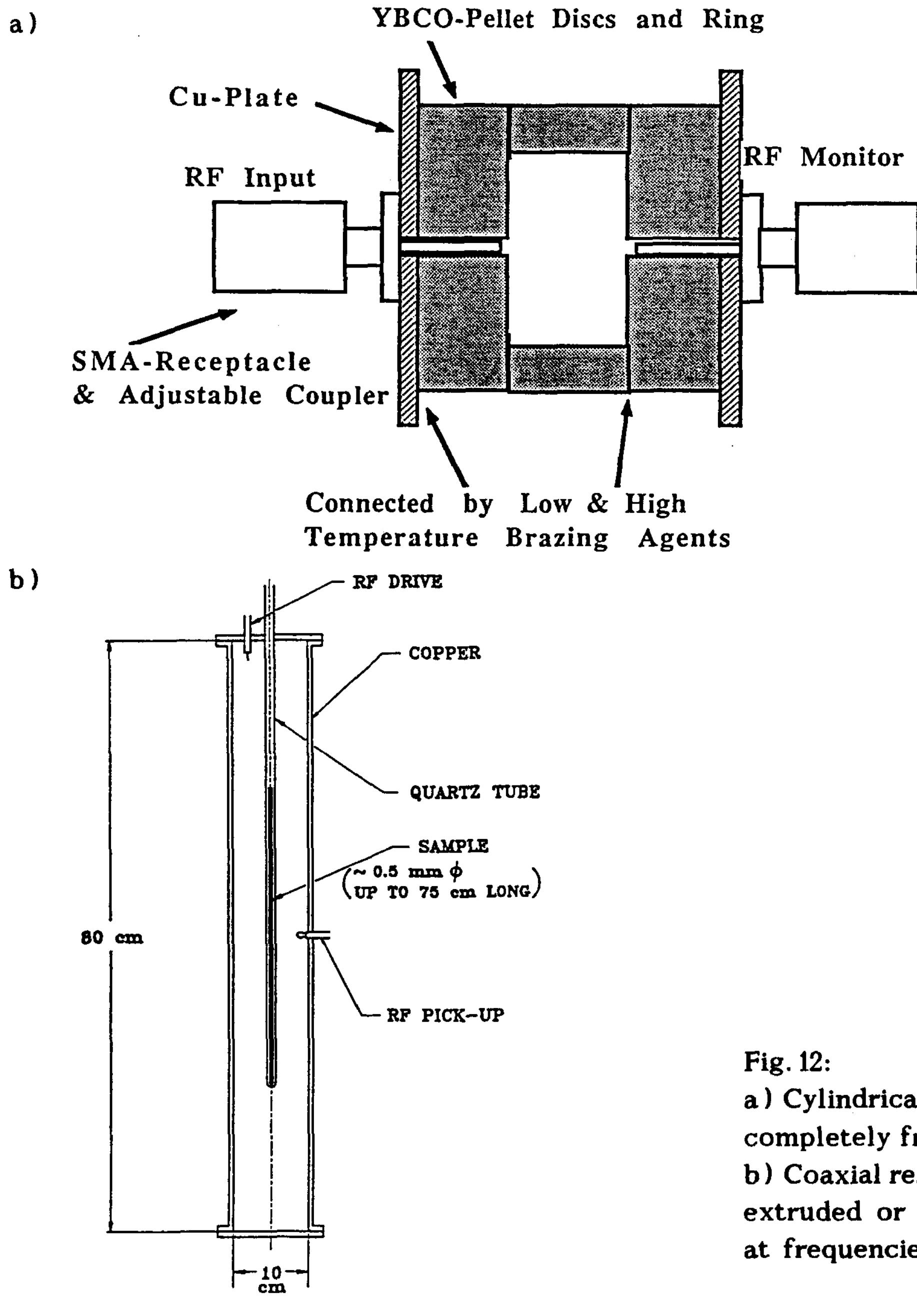



Fig. 12:
a) Cylindrical pill-box cavity built completely from $YBa_2Cu_3O_7$ [78].
b) Coaxial resonator for testing of extruded or coated wire samples at frequencies below 1 GHz [79].

For polycrystalline bulk ceramic and thick films, the full scope of cavity measurement techniques can be applied of course. Typical examples for cavities built completely or at least in their major part from oxide superconductors are shown in Fig. 12. Such cavities are most sensitive for the temperature, frequency and field dependence of the surface resistance but require the highest fabrication effort, too. At frequencies above 3 GHz, pill-box cavities assembled from rings and discs are favoured, which are connected by specially developed brazing agents to achieve high Q values in the $TM_{010}$ mode [78]. At lower frequencies, coaxial cavities using wire samples, which are directly cooled in a quartz tube, are the best choice for easy multimode operation [4, 80]. As a mixture of both, lumped circuits inside shielding cavities have been designed with pairs of wires [81] or half cylinder shells [82]. For all these kinds of cavities, the use of silver parts coated with oxide superconductors should be advantageous because of mechanical and thermal stability reasons.

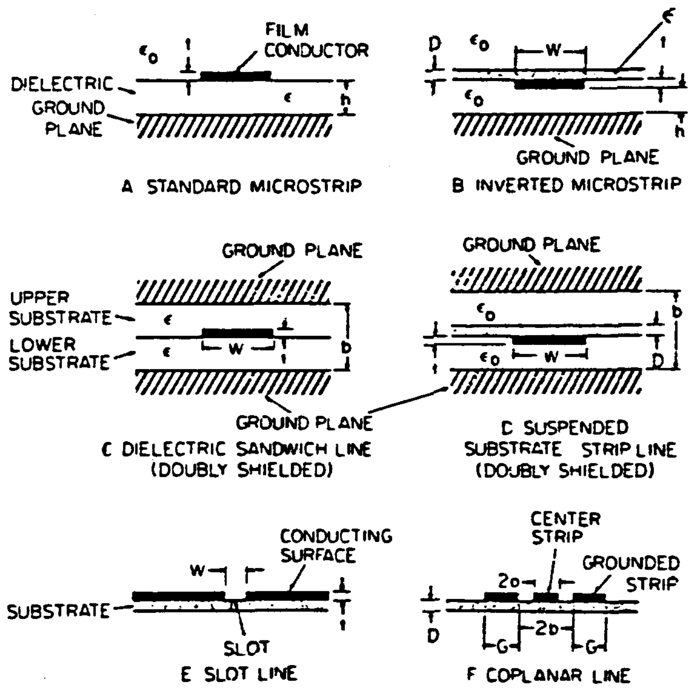



Fig. 13: Schematic configuration of microwave strips (A and B), striplines (C and D), slot line (E) and coplanar line (F) [83].

The frequency range for the test of small thin-film samples on dielectric substrates can be enormously enlarged by their patterning. In Fig. 13, six different planar microwave line configurations are displayed. Obviously, fillings and substrates with low dielectric losses are needed for high performance of superconducting devices. The attainable Q values should be lower for the open structures A, B, E and F than for the closed ones C and D due to radiation losses [83]. Using standard patterning methods, up to 30 cm long transmission lines of complex shape have been successfully produced from high quality $YBa_2Cu_3O_{7-\delta}$ films [84-88]. For some of these devices, the corresponding values of $R_s$ and $\lambda$ have been determined from the resonant frequencies and line widths by means of an appropriate analysis in which the metallic as well as the dielectric and radiation losses are considered. Moreover, such devices are very promising for electronic interconnections and miniaturized passive microwave components like resonators, filters, variable delay lines (Fig. 14a) and antennas (Fig. 14b) [89].

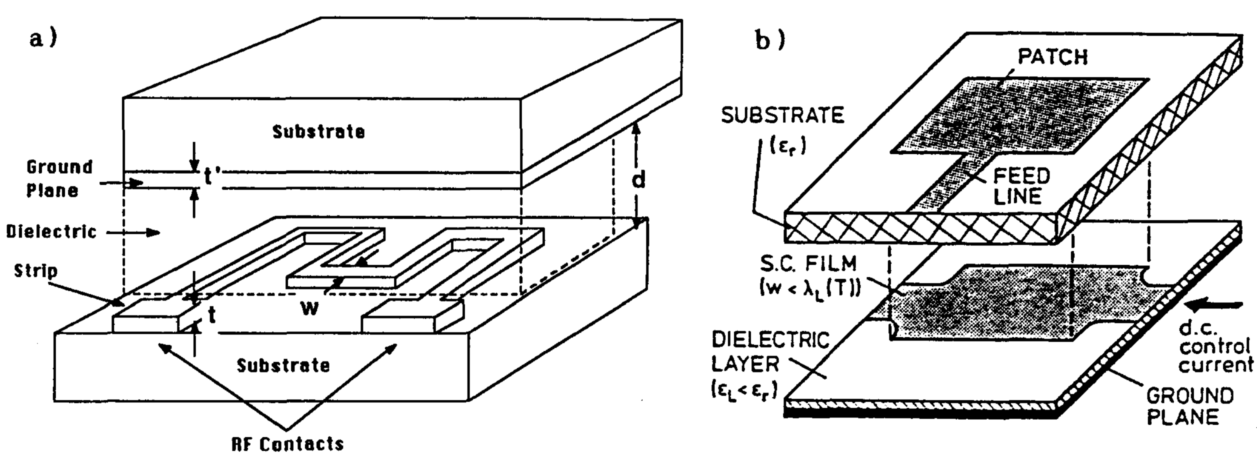



Fig. 14: Examples for applications of high-$T_c$ superconductor thin films: a) current controlled variable delay line [90]; b) electronically tunable patch antenna [91].

Tab. 4: Survey of available microwave data on the oxide superconductors $La_{2-x}Sr_xCuO_4$ (LS), $YBa_2Cu_3O_7$ (YB), $Bi_2Sr_2CaCu_2O_8$ (BSC) and $Tl_2Ba_2Ca_2Cu_3O_{10}$ (TBC) in polycrystalline (p), c-axis textured (t) and singlecrystalline (s) form.

| Laboratory | LS | YB | BSC | TBC | p | t | s | f [GHz] | References | No. |
|---|---|---|---|---|---|---|---|---|---|---|
| Wuppertal Univ. | | X | X | | X | X | X | 1.4, 3, 22, 87 | [2, 47, 50-52, 60, 67, 70, 76, 82, 91-99] | 1 |
| Argonne Nat. Lab. | | X | X | | X | | | 0.1-4, 8-12 26-40 | [4, 79, 80, 100-103] | 2 |
| Cornell Univ. | | X | | X | X | X | X | 1.5, 6 | [3, 61, 69, 104-112] | 3 |
| Boston NE Univ. | X | X | | | X | | | 8, 10 | [1, 68, 74, 113-115] | 4 |
| Los Angeles UCLA | X | X | X | X | X | | X | 102, 148 | [61, 71, 116-123] | 5 |
| Los Alamos Nat. L. | | X | X | X | X | X | X | 3, 22 | [5, 97, 98, 124-127] | 6 |
| Madison/Wi Univ. | | X | | | X | | | 3-17 | [73, 128] | 7 |
| Princeton DSRC | | X | | | X | | X | 1-14 | [61, 85, 129, 130] | 8 |
| Lincoln Lab. MIT | | X | X | | | | X | 0.6-18 | [86, 131, 132] | 9 |
| Westinghouse/Pa | | X | | | X | | | 3-10 | [83] | 10 |
| Rockwell ISC/Ca | | X | X | X | X | | X | 60 | [77, 133, 134] | 11 |
| Superc. Techn./Ca | | X | | X | X | | X | 10, 102, 148 | [61, 71, 112, 122] | 12 |
| JAERI/Japan | | X | | | X | | | 3, 7, 20 | [78, 135] | 13 |
| NTT/Japan | | X | | | X | | X | 1-8, 24 | [81, 136, 137] | 14 |
| DESY | | X | | | X | | | 0.5 | [138] | 15 |
| Stanford Univ. | | X | | | | | X | 1-25 | [84, 90, 117, 119-121, 139] | 16 |
| Houston Univ. | | X | X | | | X | | 85 | [140] | 17 |
| Teddington NPL | | X | | | X | | X | 10-18 | [72, 141-143] | 18 |
| Washington NRL | X | X | | X | X | | X | 9, 18 | [144-146] | 19 |
| Thomson CSF/LCR | | X | | | X | | | 2-10.5 | [147, 148] | 20 |
| Ford Aerospace/Ca | | X | | | X | | | 4.5 | [75] | 21 |

By means of the described microwave measurement techniques, the most interesting oxide superconductors have been investigated at numerous research laboratories as listed in Tab. 4. This review is based on about 100 publications, conference contributions, preprints and private communications on the surface impedance of oxide superconductors from 21 groups in the span of approximately two and one-half years between the first observation of rf superconductivity in $YBa_2Cu_3O_{7-\delta}$ [67] and August 1989. They cover the frequency range from 100 MHz to 150 GHz. It is remarkable that microwave data exist at all these laboratories for $YBa_2Cu_3O_{7-\delta}$ but only sporadically for other oxide superconductors. Moreover, at most laboratories polycrystalline samples were tested first before switching over to textured or singlecrystalline samples. The main reason for this development consists in the easy single-phase fabrication of bulk $YBa_2Cu_3O_{7-\delta}$, which is mostly performed in house. In comparison, textured or singlecrystalline samples of high quality have been produced only at a few of these laboratories.

## RESULTS AND DISCUSSION FOR $R_s(T, f, H_{rf})$ AND $\lambda(T)$

Because of the large number of microwave data for the oxide superconductors, it is rather confusing to report about all of them in detail. Therefore, exemplary results for common trends will be discussed first in this chapter. The anomalous temperature dependence of the surface resistance $R_s$ is demonstrated by systematic test series on polycrystalline bulk ceramic or thick films of $YBa_2Cu_3O_7$ and initial results for thick films of $Bi_2Sr_2CaCu_2O_8$ and thin films of $Tl_2Ba_2Ca_2Cu_3O_{10}$. The frequency dependence of $R_s$ obtained for such granular or multi-oriented samples follows as measured on a single $YBa_2Cu_3O_7$ thin film over a wide range. Then the best results achieved so far reproducibly for $R_s$ and $\lambda$ with singlecrystalline $YBa_2Cu_3O_{7-\delta}$ thin films on $SrTiO_3$ and $LaAlO_3$ substrates by laser ablation are presented and compared to those of high quality single crystals. In order to get an overview of the present state of the art, only the lowest $R_s$ values measured at all laboratories on different types of $YBa_2Cu_3O_{7-\delta}$ samples will be compared with respect to the operating temperature and frequency. Finally, some results on the rf magnetic field $H_{rf}$ dependence of $R_s$ will be given.

The systematically achieved improvement of polycrystalline $YBa_2Cu_3O_{7-\delta}$ samples is shown in Fig. 15. Curves 1 and 2 have been measured on the same pellet after a total sintering time of 125 h and 240 h, respectively. The corresponding decrease of $R_s$ at all temperatures has been reproduced quite often and is attributed to the homogenization of the pellet and improved intergrain contacts. However, sintering times of more than 300 h do not lead to further improvements [70]. Additional ball milling steps during the powder calcination yield after a sintering time of 282 h bulk samples with a steeper microwave transition (curve 3) but higher residual resistance than curve 2. Therefore, a better microstructure with a high degree of phase purity seems to be essential for a small transition width, while impurities seem to be at least partially responsible for the residual losses as it is the case for classical superconductors. Attempts to combine both advantages finally succeed somewhat as shown in curve 4. A further reduction of $R_s$ at temperatures below 85 K can be achieved much easier with c-axis textured thick films (curve 5), which have been electrophoretically deposited in a high magnetic field. Similar results have been obtained with magnetically aligned bulk samples [110, 112], which show a significant anisotropy of $R_s$ depending on the orientation of the rf currents

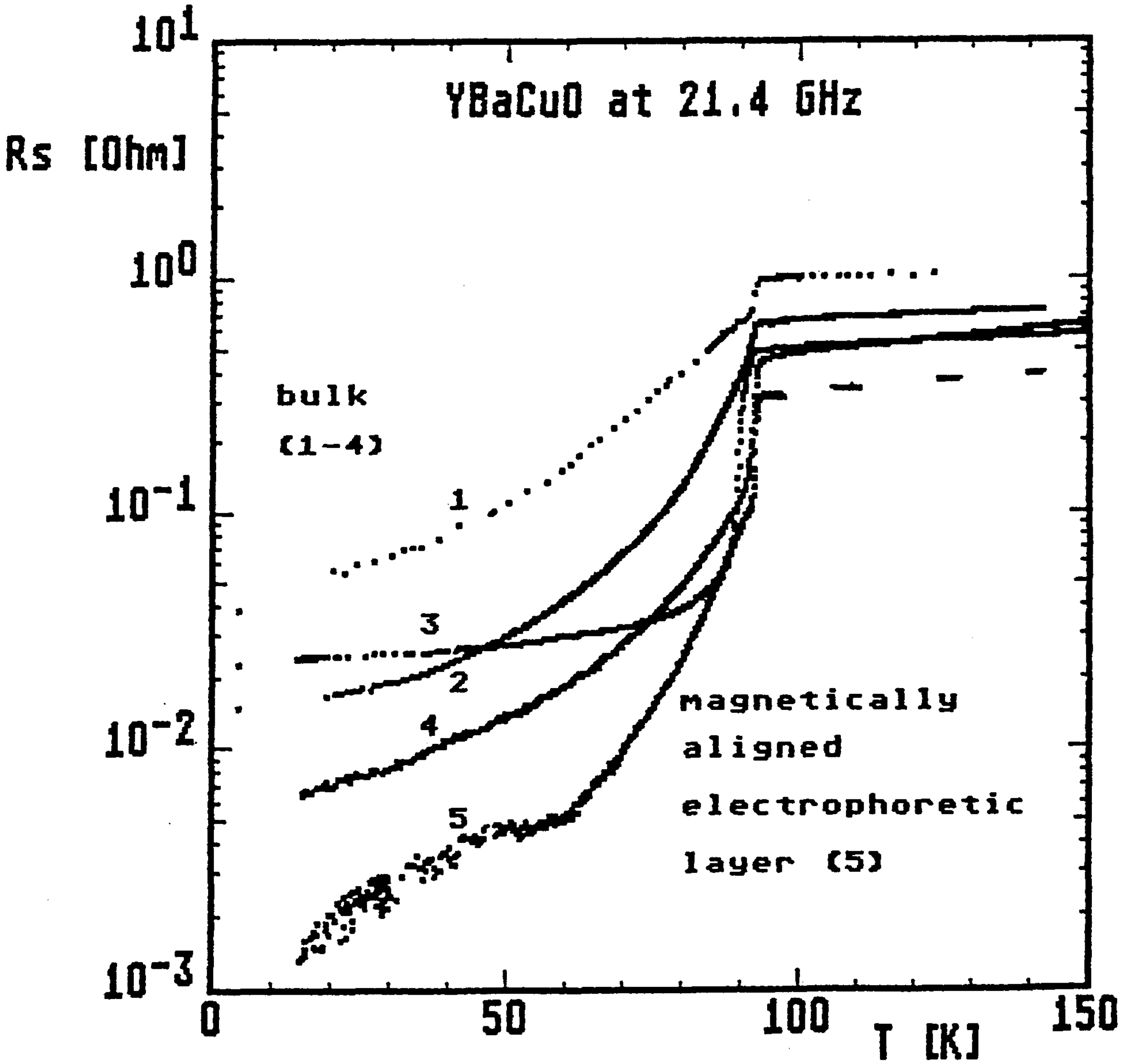
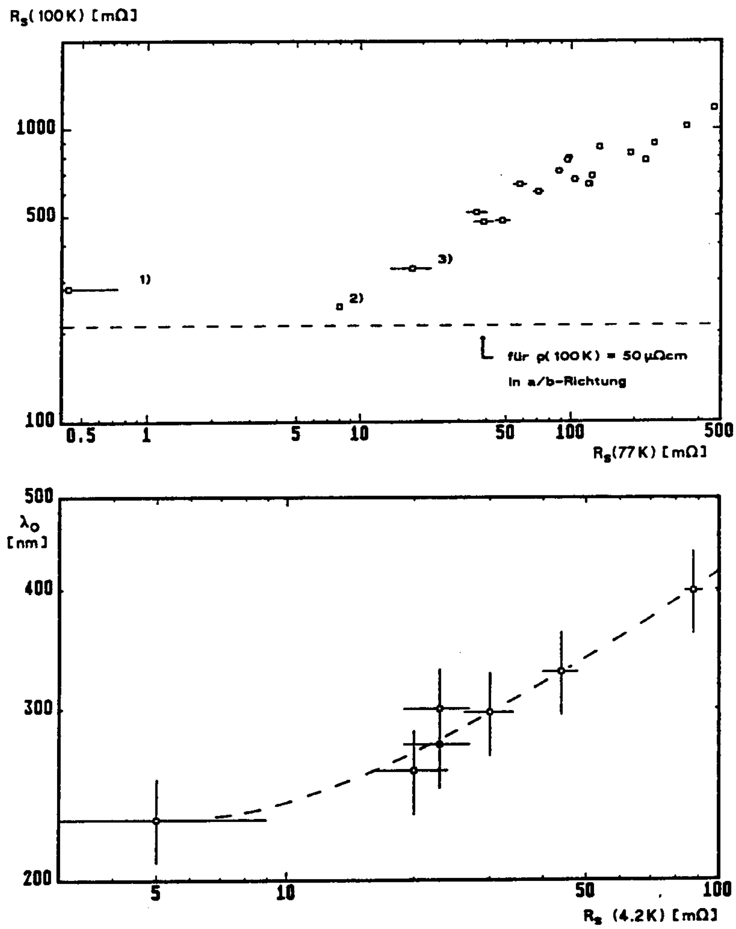



Fig. 15: Temperature dependence of the surface resistance of $YBa_2Cu_3O_{7-\delta}$ at 21.4 GHz for differently prepared pellets (1-4) [47,99] and for a c-axis textured thick film on a silver disc (5) [50,51].

with respect to the c-axis. Both results confirm the importance of in-plane currents and good intergrain contacts for low residual losses. Nonetheless, all of these polycrystalline $YBa_2Cu_3O_{7-\delta}$ samples provide much higher residual losses and broader transitions than classical supercondcutors. The major role of the degree of orientation for the performance of oxide superconductors becomes even more evident in Fig. 16. The clear correlation between $R_s$(100 K) and $R_s$(77 K) for bulk ceramic and textured thick films in Fig. 16a indicates superior quality for increasing in-plane current contribution. Moreover, it is tempting to postulate from this correlation and an intrinsic limit of about 200 mΩ for $R_s$(100 K) at 21.4 GHz a lower bound of a few mΩ for $R_s$(77 K) of granular $YBa_2Cu_3O_{7-\delta}$, which might be avoidable only with singlecrystalline films. Another clear correlation exists between the $\lambda(0)$ and $R_s$(4.2 K) values of bulk samples as shown in Fig. 16b. While the minimum $\lambda(0)$ is of course limited by the intrinsic value of about 140 nm for currents in the ab-plane, larger values are expected due to averaging over all grain orientations (see Tab. 2) and due to granularity (4). According to (6), the observed linear increase of $R_s(4.2\,K)$ with $\lambda(0)$ for medium quality bulk samples can be explained by a dominant influence of weak links on their residual losses.




a)

b)

Fig. 16:
Correlations between microwave properties of bulk $YBa_2Cu_3O_{7-\delta}$ at 21.4 GHz: a) surface resistance of normal versus superconducting state, also for epitaxially grown thin films (1), single crystals (2) and c-axis textured thick films (3), b) penetration depth $\lambda(0)$ versus residual $R_s(4.2\,K)$ [99].

In comparison to $YBa_2Cu_3O_{7-\delta}$, polycrystalline $Bi_2Sr_2CaCu_2O_8$ samples yield even broader microwave transitions and higher residual losses especially for surplus copper content [93]. This is demonstrated by the typical results shown in Fig. 17a. Although these thick films are highly c-axis textured perpendicular to the surface [100], their microwave transition widths of more than 5 K and their $R_s(4.2\,K)$ values of about 7 mΩ at 3 GHz and 30 mΩ at 10 GHz are much worse than those of $YBa_2Cu_3O_{7-\delta}$. Nevertheless, the $R_s$ data in Fig. 17a confirm above 8 GHz roughly the expected frequency dependencies, i.e. square root behavior above $T_c$ turning into a more quadratic like behavior below $T_c$. Similar high $R_s$ values of $Bi_2Sr_2CaCu_2O_8$ have been reported for bulk pellets tested at 3 GHz [125] and 21 GHz [93] as well as for single crystals tested at 150 GHz [119]. Therefore, the lack of phase purity seems to cause high microwave losses in the Bi superconductors. In contrast, first results with bulk [146] and thin-film samples [71] of the Tl superconductor provide $R_s(77\,K)$ values which are much lower than those of copper and already competitive to those of $YBa_2Cu_3O_{7-\delta}$ up to frequencies around 20 GHz. It should be mentioned, however, that the microwave transition curves for $Tl_2Ba_2Ca_2Cu_3O_{10}$ samples are still very broad as shown in Fig. 17b, while those for $Tl_2Ba_2CaCu_2O_8$ thin films are similar broad as in Fig. 17a but with an onset at 105 K [122]. Further investigations are necessary to clarify if sufficient phase purity can be achieved for the Tl superconductors.

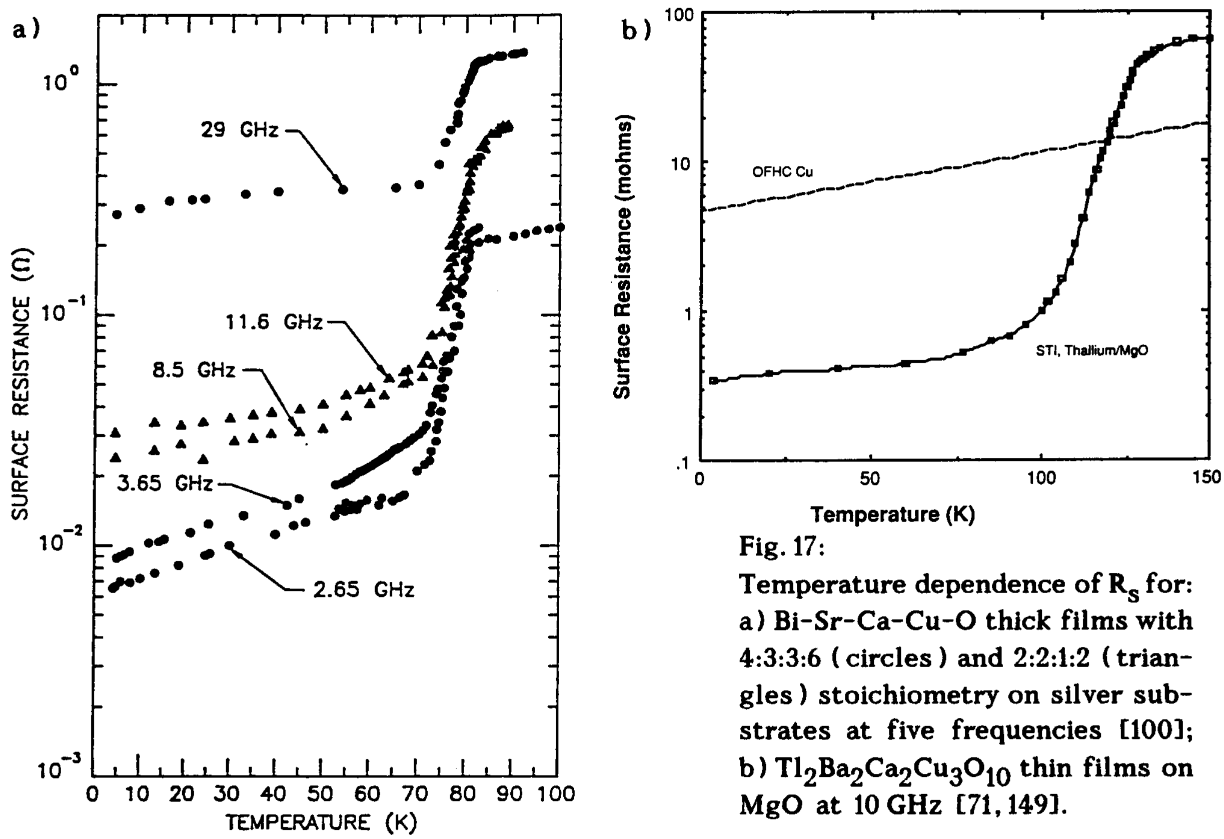



Fig. 17:
Temperature dependence of $R_s$ for:
a) Bi-Sr-Ca-Cu-O thick films with 4:3:3:6 (circles) and 2:2:1:2 (triangles) stoichiometry on silver substrates at five frequencies [100];
b) $Tl_2Ba_2Ca_2Cu_3O_{10}$ thin films on MgO at 10 GHz [71, 149].

For the determination of the intrinsic frequency dependence of $R_s$, measurements on the same sample over a large frequency range are required. Coaxial cavities like the one shown in Fig. 12b are best suited to perform this with bulk samples at low frequencies. As the result of such a test series on a $YBa_2Cu_3O_{7-\delta}$ wire sample, between 200 MHz and 1 GHz approximately a quadratic increase of $R_s$ with frequency has been observed for temperatures between 20 K and 90 K despite of the unusual $R_s(T)$ dependence [101]. Experiments with a thick film sample on a movable piston in a cylindrical cavity have given for the $R_s(\omega)$ power-law between 7 GHz and 17 GHz fit exponents of 2.42±0.6 at 77 K and 2.7±1.2 at 4.2 K [73]. Higher accuracy in the lower GHz range has been obtained with stripline [131] or microstrip [128] resonators made from polycrystalline films, which result at 4.2 K in $R_s(\omega)$ exponents close to 2. Similar measurements on a high quality thin film, which has been epitaxially grown on $LaAlO_3$ by laser ablation and patterned by standard photolithography, have yielded at 79 K between 1 GHz and 11 GHz an average $R_s(\omega)$ exponent of 1.6 [85]. In the upper GHz range, an alternative way has been pursued to determine the intrinsic $R_s(\omega)$ dependence of plane samples with test configurations like those in Fig. 11. A large sample of 25 mm diameter is first measured at 22 GHz and then cut into smaller pieces for tests at 86 GHz and 148 GHz. The result of such a test series on a multi-oriented thin film sputtered on $LaGaO_3$ is shown in Fig. 18. From these data, $R_s(\omega)$ exponents of 2.06±0.14 at 70 K and 2.02±0.47 at 30 K have been extracted [97]. In summary of all these measurements with different techniques and quality of $YBa_2Cu_3O_{7-\delta}$ samples, the quadratic frequency dependence of $R_s$ expected for homogenous superconductors according to (11) and (13) as well as for granular ones according to (6) is confirmed between 100 MHz and 150 GHz at low field levels. As long as the intrinsic absolute values of $R_s(\omega)$ are still unknown, however, the observed frequency dependence of $R_s$ seems to reflect rather the granularity of the samples.

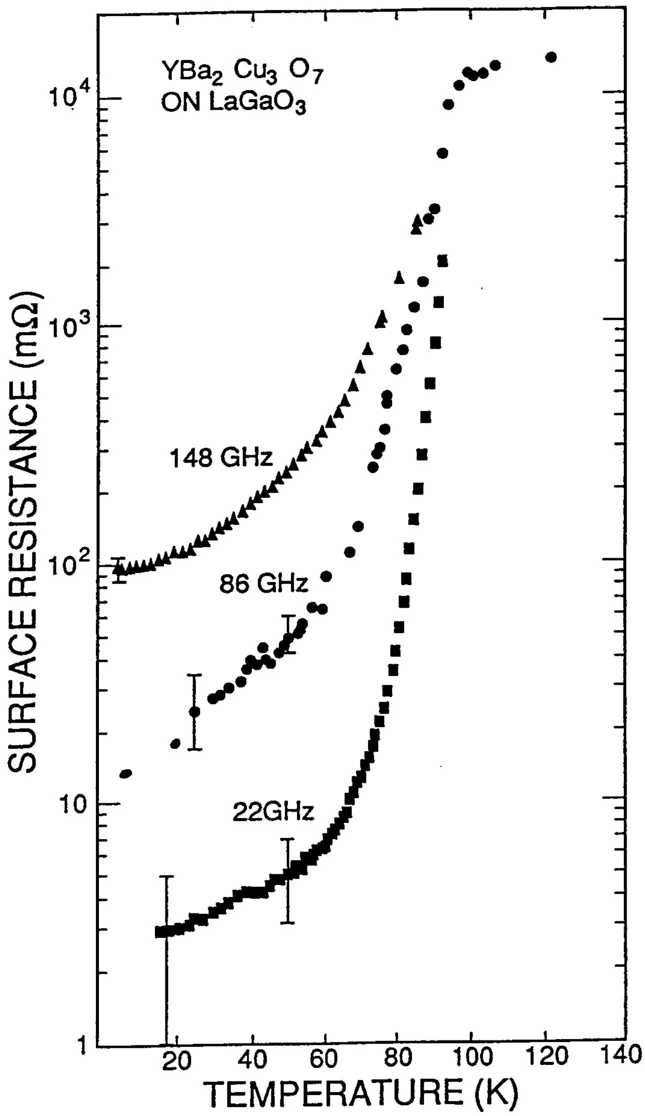
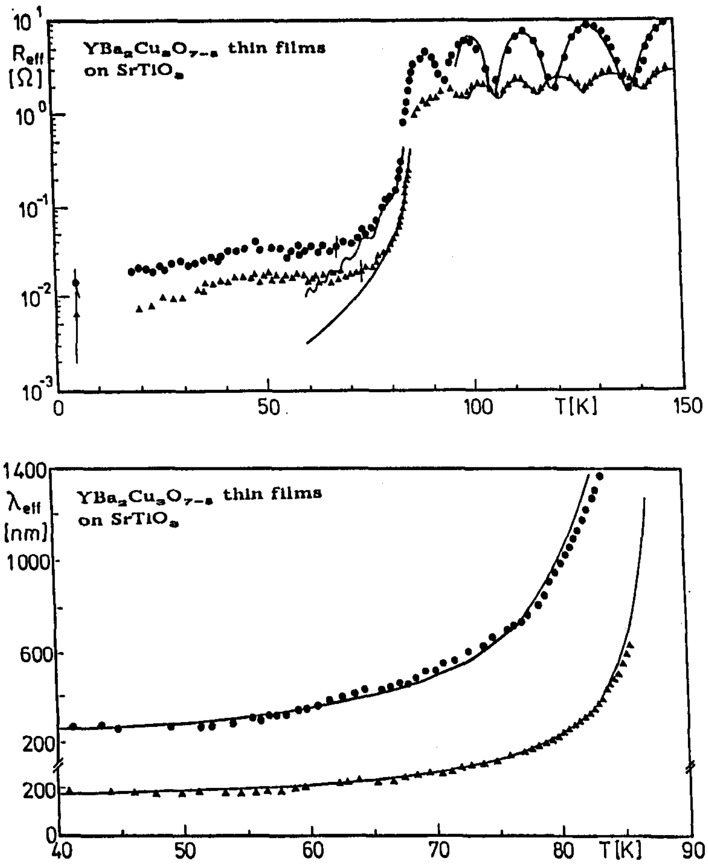



Fig. 18:
Temperature dependence of $R_s$ for a multi-oriented $YBa_2Cu_3O_7$ thin film on $LaGaO_3$ measured at Los Alamos (22 GHz), Wuppertal (86 GHz) and Los Angeles (148 GHz) [97].

The best chances to overcome the effects caused by the anisotropy and granularity of the high-$T_c$ oxide superconductors exist at present for singlecrystalline $YBa_2Cu_3O_7$ samples of high phase purity. Since about one year, c-axis oriented $YBa_2Cu_3O_{7-\delta}$ thin films of 1 $cm^2$ size are available, which have been epitaxially grown on $SrTiO_3$ substrates by the laser ablation technique. In Fig. 19, the surface resistance and penetration depth for two of such films with different thickness are displayed. The oscillations of $R_s$ in Fig. 19a are caused by standing waves in the substrate and are correlated to the strong temperature dependence of the permittivity of $SrTiO_3$ (see Tab. 3). This has been verified by numerical calculations of the effective surface impedance of the sandwich structure, which are based on multiple reflections of plane waves at all boundaries as described in detail elsewhere [96]. Depending on the ratio of the film thickness and $\lambda$, the partial penetration of the electromagnetic fields into the substrate leads to enlarged effective $R_{eff}$ and $\lambda_{eff}$ values. The excellent fit of the surface impedance data by the calculated effective values for the known film thickness allows the extraction of the true $R_s(T)$ and $\lambda(T)$ values of the films [76]. In the normal conducting regime, typical resistivities $\rho$ of about 95 µΩcm at 100 K and 290 µΩcm at 300 K have been determined according to (1) from the $R_s(T)$ data of these films, which include loss contributions from the still present surface imperfections, too. In the superconducting regime, both films exhibit in accordance with (11) a sharp microwave transition to an $R_s(77\,K)$ value of $(8\pm2)\,m\Omega$. Below 70 K, $R_s$ is dominated by a weakly temperature dependent residual surface resistance, which increases for thicker films most probably due to the observed higher density of imperfections. The measured $\lambda(T)$ data in Fig. 19b can be fitted best in the frame of the BCS theory in the weak coupling limit [150]. For both films, the resultant $\lambda(0)$ value of $(160\pm20)$ nm is in good agreement with literature data for in-plane currents (see Tab. 2). Measurements on a coplanar transmission line resonator patterned from such a film on MgO have given at 9 GHz and 77 K an overall quality factor of 1300 which is 14 times higher than that of an equivalent copper resonator and which comes close to the calculated Q, if a quadratic scaling of $R_s(\omega)$ is assumed [88]. Much improved Q values of about 10000 are expected for the same structure, if high quality $YBa_2Cu_3O_7$ films on $LaAlO_3$ substrates could be used.




a)

b)

Fig. 19:
Temperature dependence of $R_{eff}$ (a) and $\lambda_{eff}$ (b) at 86-87 GHz for two c-axis oriented $YBa_2Cu_3O_{7-\delta}$ films on $SrTiO_3$ of 170 nm (circles) and 440 nm (triangles) thickness. The full lines result from the numerical calculations [76]. Please note the broken $\lambda_{eff}$ scale.

Recently, $YBa_2Cu_3O_7$ thin films with low microwave losses have been deposited onto $LaAlO_3$ by in-situ laser ablation [111]. Contrary to similar films on $SrTiO_3$, these films can be measured like single crystals inside superconducting host cavities as shown in Fig. 10 because of the much reduced dielectric loss tangent of the substrate (see Tab. 3). The $R_s(T)$ curves achieved at 6 GHz for two of such films are compared to those of a high quality single crystal in Fig. 20. Obviously, both films show a narrower microwave transition at higher $T_c$ values but a similar residual surface resistance, which comes already close to the sensitivity limit of the apparatus. Additional measurements on the same films at 10 GHz and 100 GHz as well as on a patterned film between 2 GHz and 12 GHz have confirmed their superior quality and the expected quadratic frequency

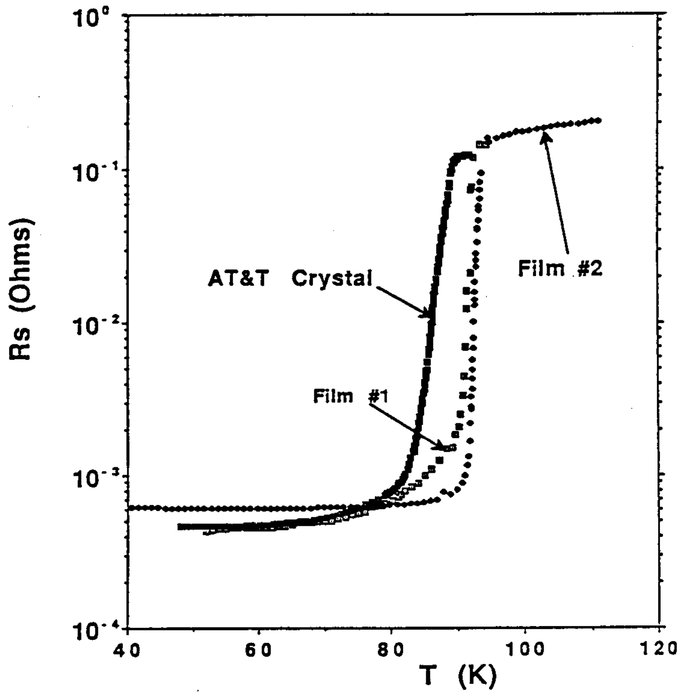



Fig. 20:
Temperature dependence of $R_s$ at 6 GHz for two $YBa_2Cu_3O_{7-\delta}$ thin films on $LaAlO_3$ and for a single crystal of high quality [111].

dependence of $R_s$(77 K) [61]. Nevertheless, the high residual losses of these films still prohibit the determination of the reduced energy gap. This is demonstrated in Fig. 21, where the measured temperature dependence of $R_s$ is plotted according to (13) as a semi-logarithmic function of $T_c/T$. Although the drop of $R_s$ near $T_c$ is as sharp as for classical superconductors like $Nb_3Sn$, the expected exponential decrease cannot be revealed so far for any thin film or bulk samples of $YBa_2Cu_3O_7$ due to the unknown origin and temperature dependence of the high residual surface resistance. Moreover, higher operating frequencies are favourable for the determination of $\Delta_0/kT_c$ only if the residual losses increase less than quadratically with the frequency.

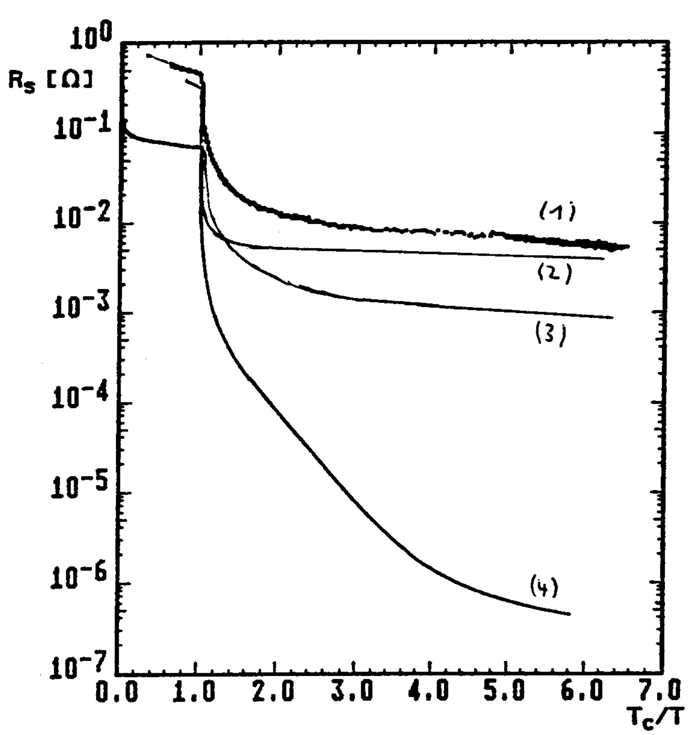



Fig. 21:
Surface resistance versus reduced temperature for bulk (1) [99], laser ablated (2) [111] and coevaporated samples (3) [127] of $YBa_2Cu_3O_7$ in comparison to that of $Nb_3Sn$ (4) [151]. While curves (1), (3) and (4) have been measured directly at 22 GHz, curve (2) has been scaled quadratically from 6 GHz data.

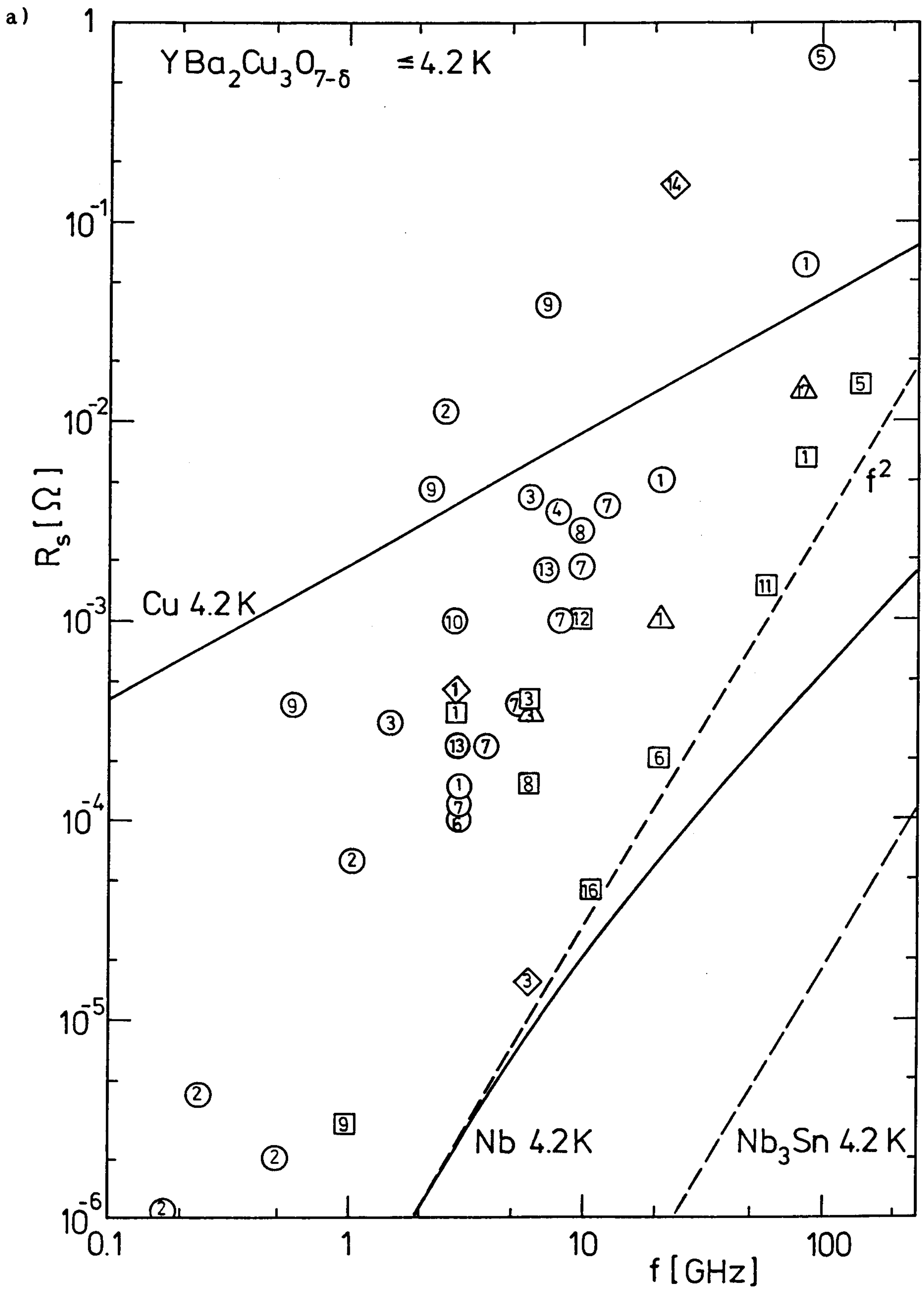
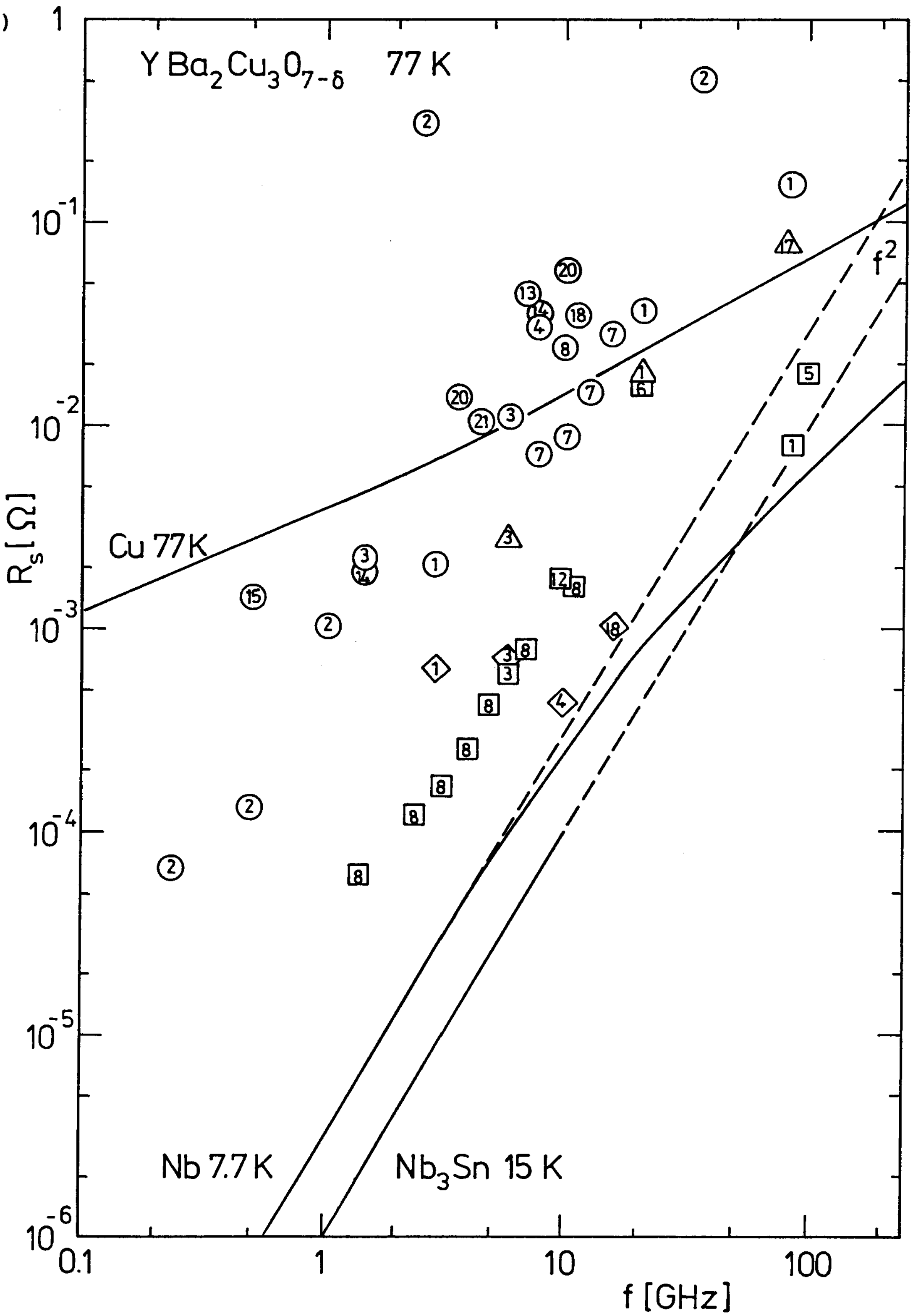



Fig. 22: Frequency dependence of $R_s$(4.2 K) (a) and $R_s$(77 K) (b) for untextured (circles) and c-axis textured (triangles) polycrystalline bulk or thick film samples as well as for epitaxially grown thin films (squares) and single crystal platelets (rhombuses) of $YBa_2Cu_3O_{7-\delta}$ at all laboratories listed in Tab. 4 (numbers in symbols). The lines for copper, niobium and $Nb_3Sn$ are given for comparison.

b)




At this point it is helpful to review the lowest $R_s$ values of $YBa_2Cu_3O_{7-\delta}$ achieved for low magnetic surface field levels at all laboratories listed in Tab. 4. In Fig. 22, this is done seperately for 4.2 K and 77 K as a function of frequency. Included are only the best results from every laboratory for the main categories of samples and for different operating frequencies. At first sight, the scattering of these data beyond the expected quadratic $R_s(\omega)$ scaling even within the same category reflects the different quality of the samples. Moreover, there is a clear evidence of reduced losses for higher degree of c-axis orientation, which underlines the influence of anisotropy and granularity on the measured $R_s$ values at both temperatures. In this respect it is most remarkable that the record data for $R_s$(4.2 K) of singlecrystalline samples at six different laboratories coincide approximately with the dashed $f^2$ line in Fig. 22a, which is very close to the prediction of ( 8 ) derived for an intrinsic strong granularity of $YBa_2Cu_3O_7$. On the other hand it is difficult to understand, how such losses could occur in a perfectly c-axis oriented thin film, which provides in-plane currents only. Therefore, improved epitaxially grown thin-film samples with less imperfections are necessary to get a better judgement about the relevance of this limit. The only positive aspect of such a limit would be that it should stay at 77 K in the same order of magnitude, because only slight changes of the parameters $\Delta$, $R_{in}$ and $I_{i0}$ in eqs. ( 2 ) to ( 6 ) are expected up to this temperature. This consideration could be an explanation for the rather flat $R_s(T)$ curves of very good samples ( Fig. 20 ) at low temperatures. In contrast, polycrystalline samples with higher residual losses due to weak links at grain boundaries or phase impurities show below $0.9\,T_c$ a strong temperature dependence of $R_s$ ( Fig. 15 ). It is interesting to note that the best $R_s$(77 K) values of singlecrystalline $YBa_2Cu_3O_{7-\delta}$ come already close to the $R_s(\omega)$ data of classical superconductors like Nb [152] and $Nb_3Sn$ [153] at the same reduced temperature of 7.7 K and 15 K, respectively ( see Fig. 22b ). If one applies the BCS theory to $YBa_2Cu_3O_7$, however, a much lower $R_s$(77 K) value of about 0.2 mΩ at 87 GHz results from [66] for the material paramaters in Tab. 2 ( assumed $\Delta_0/kT_c \approx 2$ ) due to the higher gap frequency. From a more practical point of view, the comparison of the present data with the $R_s(\omega)$ curves for copper in Fig. 22 is rather encouraging for applications of polycrystalline $YBa_2Cu_3O_7$ up to about 10 GHz and of singlecrystalline $YBa_2Cu_3O_7$ up to THz frequencies at temperatures between 20 K and 80 K and at low field levels. Moreover, the high-$T_c$ oxide superconductors are potentially useful for applications even at lower temperatures if higher operating frequencies or field levels than with classical superconductors could be achieved.

The systematic investigation of the rf magnetic field dependence of $R_s$ is complicated by the high power needs and the resultant cooling problems for samples of poor quality. Meanwhile, there are some interesting data for polycrystalline as well as for single-crystalline $YBa_2Cu_3O_7$. At Argonne, the direct cooling of wire samples with LHe or $LN_2$ inside coaxial resonators ( Fig. 12b ) has enabled to measure their microwave response up to surface field levels of nearly 100 mT ( ≙ 1000 Oe ) as shown in Fig. 23. $R_s$(77 K) starts to increase already at $H_s$ levels as low as 1 μT, especially if a residual loss contribution is substracted which depends not on $H_s$ but quadratically on frequency. Between 100 μT and 0.3 mT for 888 MHz or 2 mT for 190 MHz, a clear rise of $R_s$(77 K) is observed as expected ( 9 ) for granular material. Although it scales slightly more than linearly with both $H_s$ and f, from the average slope of about 0.6 mΩ/Oe at 430 MHz and ( 9 ) a critical current density $J_c$ of $10^4\,A/cm^2$ can be derived. At higher field levels, a plateau occurs which amounts to about 5 % of the corresponding $R_s$ value just above $T_c$. Therefore, rf superconductivity still prevails in 95 % of the sample up to the highest field of 64 mT. Additional measurements of $R_s(H_s)$ at 4.2 K have given similar increasing curves and plateau heights but slightly reduced slopes as those at 77 K [101]. For bulk pellets in pulsed power operation, linear slopes for $R_s(H_s)$ of about 1 mΩ/Oe have been obtained at 3 GHz [50]. All of these results can be explained by the assumption that the weak

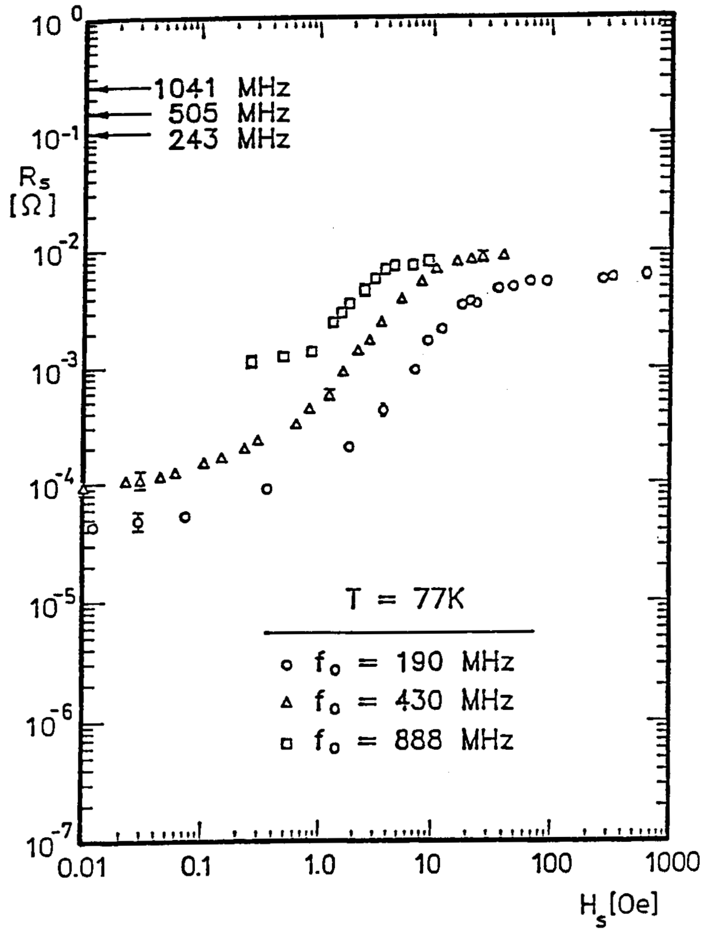



**Fig. 23:**
**Dependence of $R_S$ on the magnetic surface field $H_S$ for bulk samples of $YBa_2Cu_3O_7$ at 77 K and at three frequencies. The arrows indicate the corresponding $R_S$ values just above $T_C$ [101].**

**links switch for increasing fields successively into a resistive state. It is remarkable, however, that the $J_C$ values derived from the $R_S(H_S)$ slopes according to (9) are much higher than those measured directly on such samples. Contrary to the results on bulk samples, no substantial increase of the residual losses is observed over a wide field**

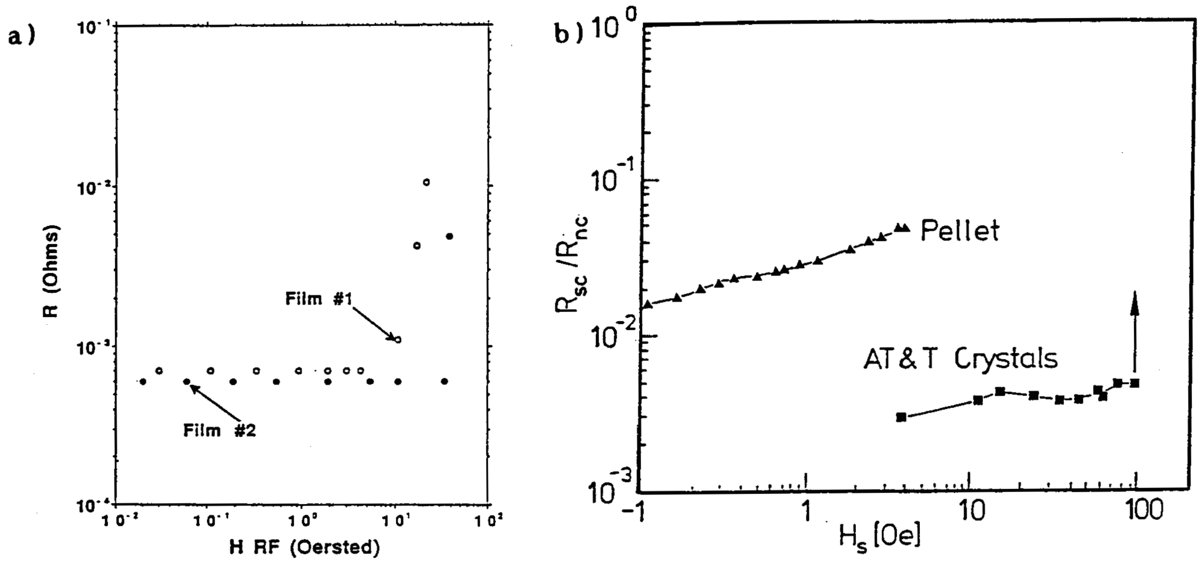



**Fig. 24: $R_S$ versus magnetic surface field $H_S$ at 6 GHz (a) for epitaxially grown thin films [111] and (b) for single crystals [69]. The nominal cavity temperature is 4.2 K, but near the highest fields the actual temperature of the samples is at least 20 K.**

range for singlecrystalline samples as shown in Fig. 24. This result supports strongly the previous discussion about weak links as the origin for increasing losses at high fields. Depending on the quality of the sample and the cooling conditions, a rapid rise of $R_s$ occurs at less than 5 mT for thin films ( Fig. 24a ) and about 10 mT for single crystals ( Fig. 24b ). Further systematic investigations are necessary to answer the question about the ultimate breakdown fields of the high $T_c$ oxide superconductors, which are of highest importance for applications in particle accelerators as well as in microelectronics.

## CONCLUSIONS

Within the last two years, systematic improvements have been achieved for the synthesis of the high-$T_c$ oxide superconductors. Measurements of the surface impedance in the whole microwave range between 100 MHz and 150 GHz using various techniques have proven to be a sensitive means to maximize the phase purity and to minimize the granularity for different kinds of samples. Sufficient phase purity has been obtained mainly for $YBa_2Cu_3O_7$, which has been investigated most, and without too much damage also for $Bi_2Sr_2CaCu_2O_8$, $Tl_2Ba_2CaCu_2O_8$ and $Tl_2Ba_2Ca_2Cu_3O_{10}$. The granularity can be suppressed partially by the c-axis texturing of bulk samples or thick films on silver substrates in high magnetic fields and more effectively by the epitaxial growth of thin films on dielectric substrates. However, an intrinsic granularity of the high $T_c$ oxide superconductors due to the very short coherence length in the c-direction cannot be excluded at present from the existing surface resistance data. The most severe obstacle are the high residual values of the surface resistance at 77 K and 4.2 K. These scale at low fields about quadratically with frequency for all types of samples and at high fields rather linearly with frequency and field strength only in case of granular samples. Moreover, magnetic field penetration depths $\lambda(0)$ of about 160 nm for in-plane currents and of at least 230 nm for polycrystalline samples have been determined.

Nevertheless, the following future prospects for applications result from the actual knowledge and state of the art for $YBa_2Cu_3O_{7-\delta}$. Polycrystalline bulk ceramic and thick films behave like granular superconductors and will be useful only at very low field levels and microwave frequencies. Singlecrystalline thin films with c-axis orientation perpendicular to the surface behave close to $T_c$ like classical superconductors but strongly anisotropic and with a residual surface resistance at lower temperatures, which is high compared to classical superconductors but low compared to copper. Therefore, such films should be very useful for planar miniaturized microwave components as well as for microelectronics. C-axis textured thick films on silver substrates could be the rigth compromise between high performance and technical applicability at least for low field applications but hopefully also for accelerator cavities.

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